Challenges in Multiphase Reactors for More Efficient Technologies

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Multiphase Reactors for More Efficient Technologies: Role of MFS and MFE

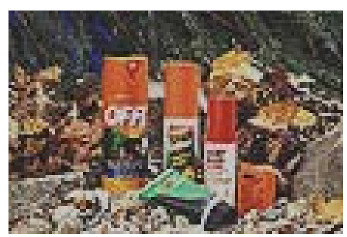
- Technology efficiency and environmental impact
- Current state of the art in process technology and multiphase reactor selection, design and scale up
- Role of multiphase flow science (MFS) in risk reduction for implementation of novel reactor technology
- Needs for effective flow models for multiphase reactive systems to improve efficiency and safety
- Suggestions for more rapid transfer of MFS into reaction engineering of multiphase systems

Key Factors Affecting the Environment and Sustainability

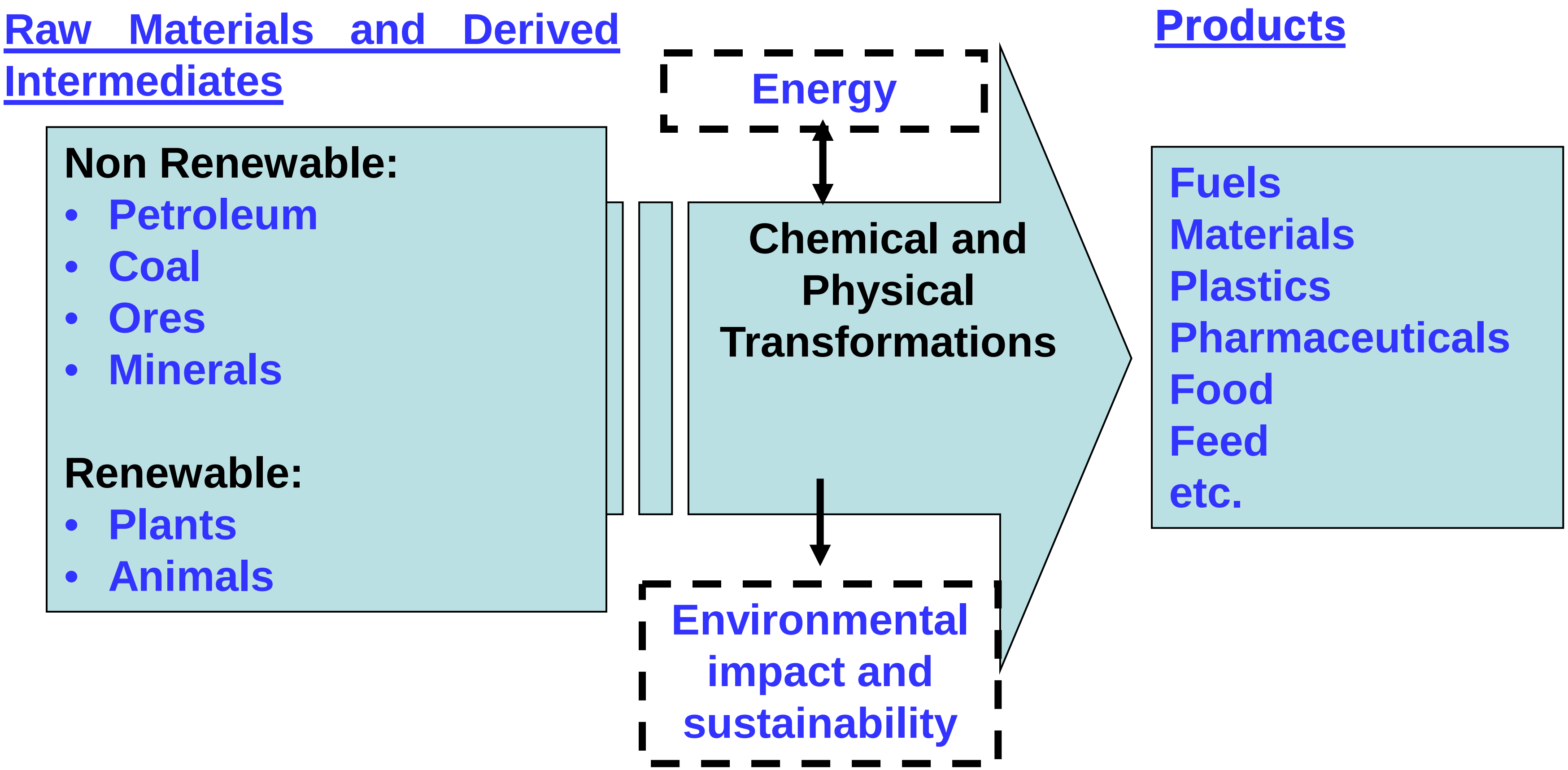
- Total population
- Lifestyle

- Recreational activities
- Manufacturing practices

- Agricultural practices
- Mining practices
- Energy utilization



Tunca, Ramachandran, Dudukovic “Role of CRE in Sustainable Development”, Sustainable Engineering Principles, M. Abraham, et. al, Ed., Elsevier (2005)

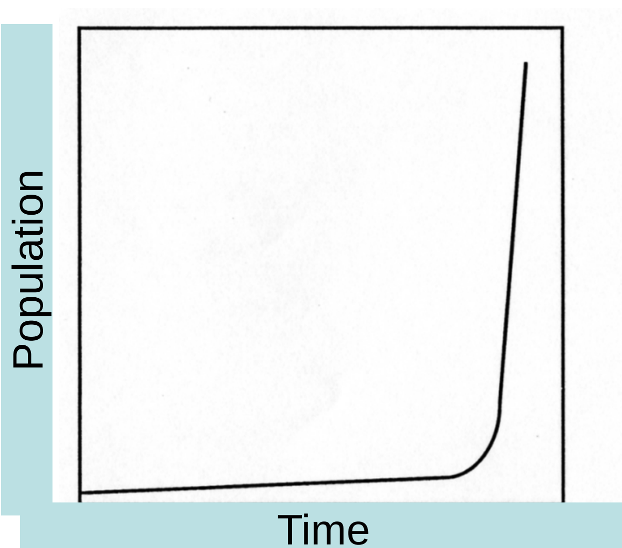


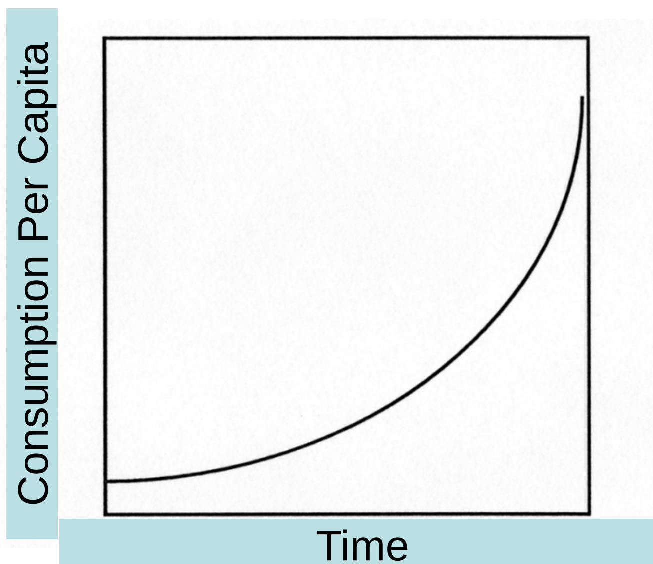
Challenges: Cleaner, sustainable processes; increased atom and energy efficiency; improved safety; ability to scale-up.

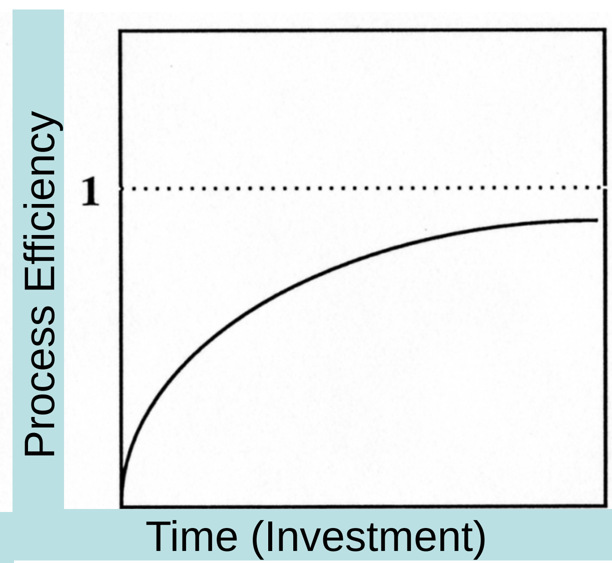
Profitability and Environmentally Benign Processing

GLOBAL VIEW

$$\underset{\text{(pollution)}}{\text{Global environmental impact}} \propto \underbrace{\text{(1-process efficiency)}}_{\text{Process inefficiency}}\ \text{(consumption per capita) (population)}$$







To raise the living standards of the poor and have a positive impact on the environment and on the world economy, while reducing global pollution, novel high efficiency processes and product manufacturing technologies are needed .

MFS and MFE have an important role to play in their development.

Also needed more conservation and recyclables oriented life style that minimizes waste and energy and materials inefficiencies.

Energy

Coal, oil, gas, nuclear power plants

Synthesis & Natural Gas Conversion

MeOH, DME, MTBE, Paraffins, Olefins, Higher alcohols, ….

Petroleum Refining

HDS, HDN, HDM, Dewaxing, Fuels, Aromatics, Olefins, ...

In USA alone

Value of Shipments:

$US 640,000 Million

Uses of Multiphase Reactor Technology

Bulk Chemicals

Aldehydes, Alcohols, Amines, Acids, Esters, LAB's, Inorg Acids, ...

Polymer and Materials Manufacture

Polycarbonates, PPO, Polyolefins, Specialty plastics; semiconductors etc

Fine Chemicals, Pharmaceuticals,Nano materials

Ag Chem , Dyes, Fragrances, Flavors, Nutraceuticals

Biomass Conversion

Syngas, Methanol, Ethanol, Oils, High Value Added Products

Environmental Remediation

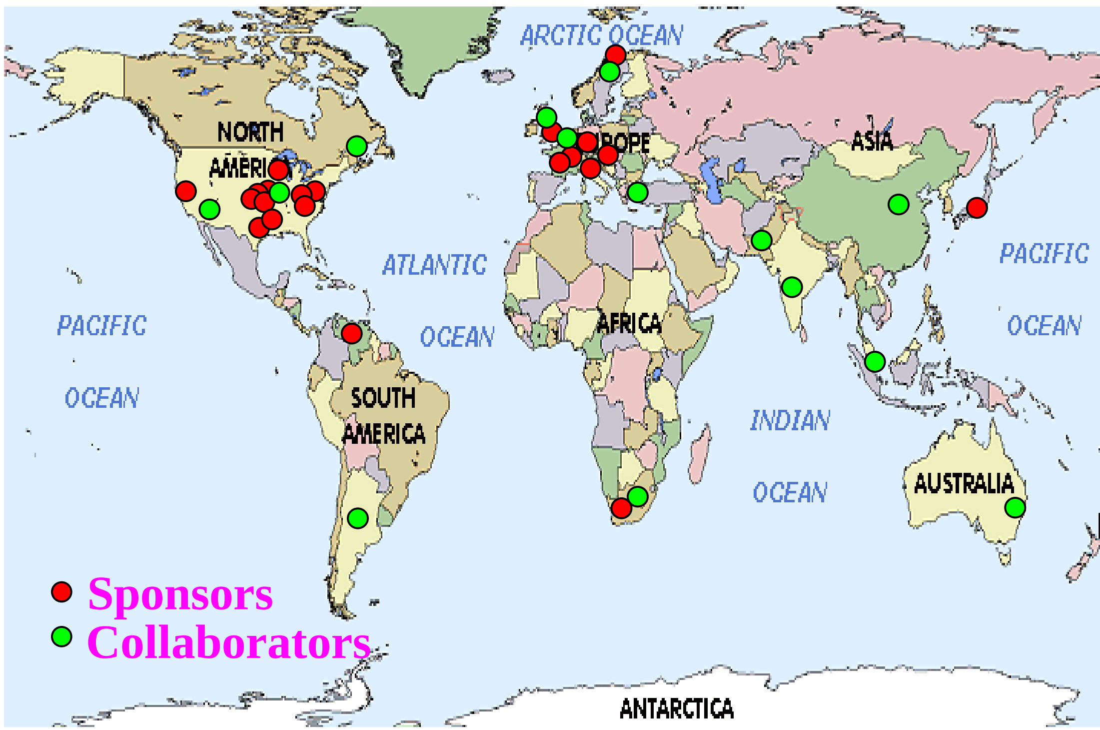
De-NOx, De-SOx, HCFC's, DPA, "Green" Processes ..

In addition: Minerals processing via hydro and pyro metallurgy

Objectives of CREL since 1974

- Education and training of students in multiphase reaction systems
- Advancement of reaction engineering methodology via research
- Transfer of state-of-the-art reaction engineering to industrial practice

CREL Sponsors and Collaborators



Industrial Sponsors

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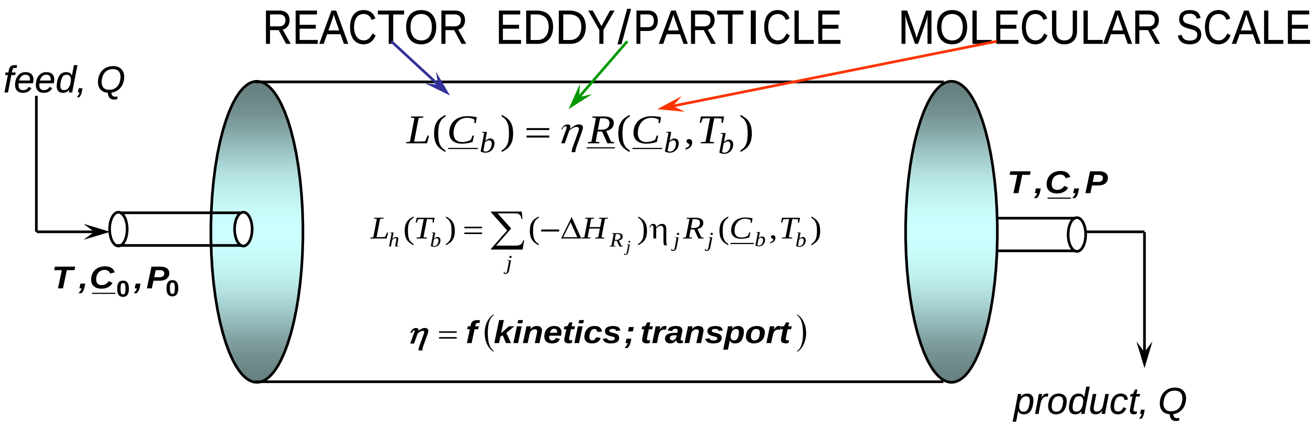
Governmental Sponsors

DOE, NSF, USDA

Multi-Scale Chemical Reaction Engineering (CRE) Methodology



REACTOR PERFORMANCE = f (input & operating variables ; rates ; mixing pattern)

Reactor choice determines plant costs; Need improved reactor selection and scale-up

10^{-10} m — 10^{-16} (s)

<u>MOLECULAR SCALE (RATE FORMS)</u>

Strictly Empirical — Mechanism Based — Elementary Steps

<u>EDDY OR PARTICLE SCALE TRANSPORT</u>

Empirical — Micromixing Models — DNS / CFD

<u>REACTOR SCALE</u>

PFR/CSTR — Axial Dispersion — Phenomenological Models — CFD

<u>PROCESS SCALE</u>

10^{2} m — 10^{4} (s)

Steady State Balances — Dynamic Models for Control & Optimization

Dudukovic, Larachi, Mills, Catalysis Reviews (2002), 44(1), 123-246

Reactor Models

- All reactor models are based on the principle of conservation of mass, species mass, energy and momentum applied to a properly selected control volume in the system:

(rate of accumulation)=

(rate of input) - (rate of output) + (rate of generation)

NOTE: The reaction rate formulation to be used in the reactor scale model must properly incorporate the key features of all the smaller scales.

The control volume size and dimensionality of the model depend on the level of knowledge of the flow field, phase distributions and exchange rates between them. Models vary from assumptions of ideal flow fields (i.e. plug flow or perfect mixing on one end to CFD descriptions of the system.

Reactors of *High Volumetric Productivity* and *High Selectivity* are needed for efficient environmentally friendly technologies.

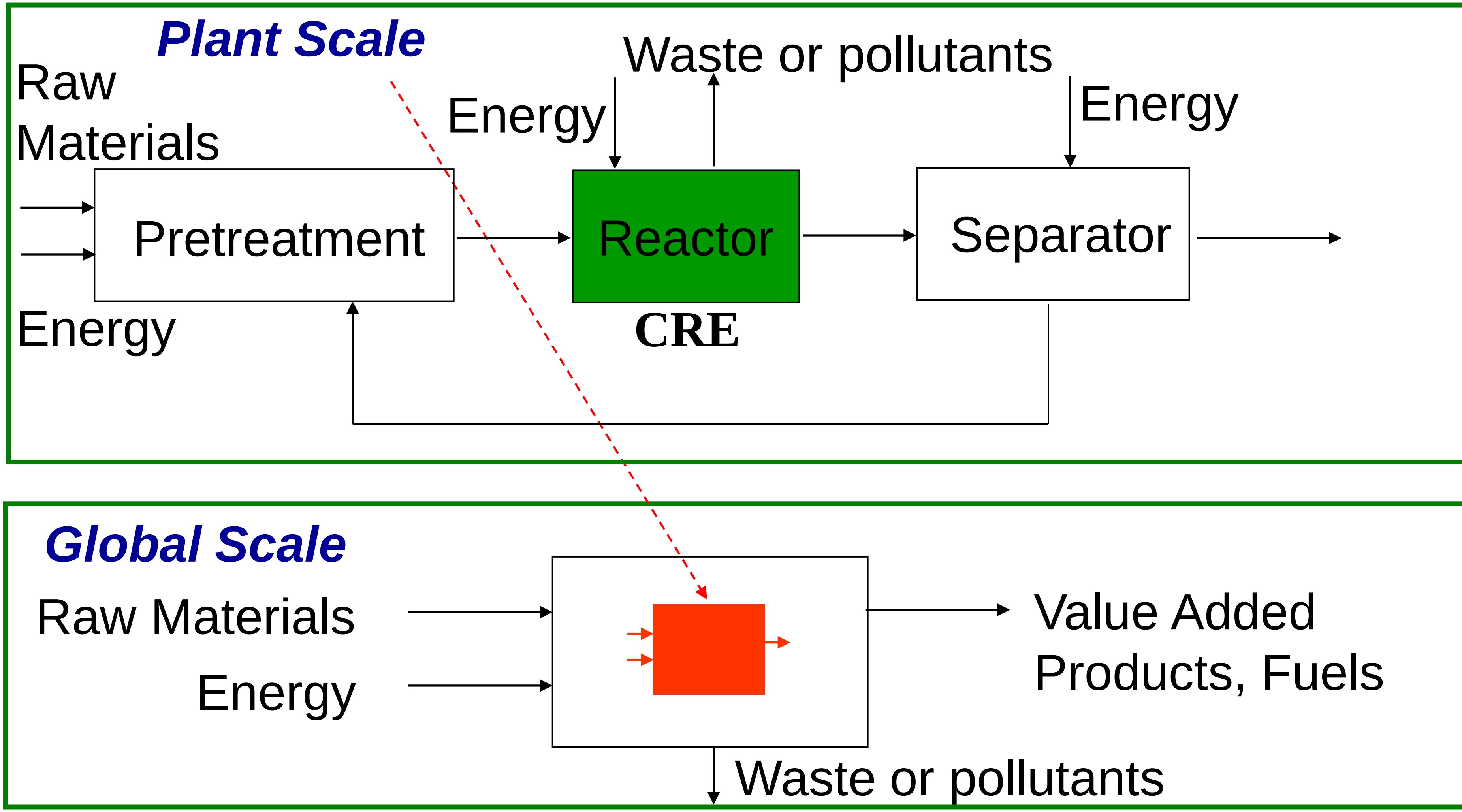
Need for Reactor Models Based on Science

- *Risk reduction of novel reactor technology or of existing reactors for new applications*
- Improved safety of existing and novel reactor types
- *Proper more accurate assessment of the environmental impact of new process technology*

Environmental Acceptability, as Measured by the E-Factor

Industry	Product tons per year	Waste/product ratio by weight
Oil refining	$10^6 – 10^8$	~ 0.1
Bulk chemicals	$10^4 – 10^6$	< 1 – 5
Fine chemicals	$10^2 – 10^4$	5 – 50
Pharmaceuticals	$10^0 - 10^3$	25 - > 100

Green Chemistry and Green Processing



Washington
WASHINGTON·UNIVERSITY·IN·ST·LOUIS
School of Engineering & Applied Science

Waste Reduction (WAR) Algorithm

Hilaly and Sikdar, Journal of the Air and Waste Management Association, 44, 1303-1308 (1994)

Available at : www.epa.gov/oppt/greenengineering/software.html

Based on Conservation Equations for mass and energy in process flow sheet.

It assesses the impact of proposed process chemistry on the environment and factors in its environmental persistence via factors on : **Acidification, Greenhouse enhancement, Ozone depletion, Photochemical oxidant formation.**

Advantages:

Important general framework based on conservation laws

Provides a metric for the environmental friendliness of a process.

Can be used to evaluate process modifications for their environmental impact

Disadvantages:

Does not directly provide any guidance on the actual origin of the waste in the process or the modifications that would minimize the waste (lacks rational cause-effect relations).

Does not guarantee correct assessment of system's performance due to low level models or due to outdated heuristics.

*: I. Halim, R. Srinivasan, Environ. Sci. Technol. 2002, 36, 1640-1648

Role of MFS in Improving the Level of Science in Multiphase Reactor Models

- Expand MFS to MFE (multiphase flow engineering)
- Create a general framework for handling reactor scale problems using subscale models that are increasingly based on fundamentals. This

  -allows selection of right reactor type for given chemistry

  -allows more accurate determination of environmental impact

  -reduces the risk of implementation of new technology

  -improves safety

  Main Challenges:

  - Create efficient framework for linking multi-scale models

  -Provide experimental validation and verification

  -Make the education on multiphase reactors and tools for handling the multi-scale reactor modeling widely available

The key function of process engineers is to transfer scientific discoveries into new technologies and practice for the benefit of mankind. This should be done based on fundamentals as much as possible and the tools for doing it should be made readily available.

Non Technical Barriers: Chase for short terms profits encourages:

Use of old 'best available technologies' which are inefficient

Use of familiar reactors and separations (contractors)

Building the plant with minimal scale-up expenditures

As a result when new chemistry is chosen one often experiments with the plant to determine 'best conditions' via statistical analysis. Very costly. Not always successful

Technical Barriers:

Manufacturing companies and engineering contractors lack expertise in multi-scale multiphase reaction engineering

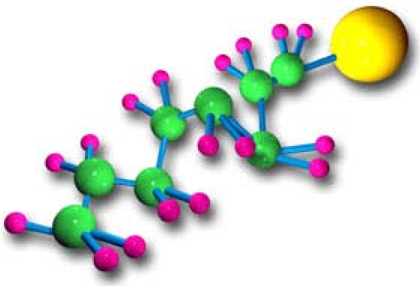
Designs are based on old correlations and integration of multi-scale concepts is missing

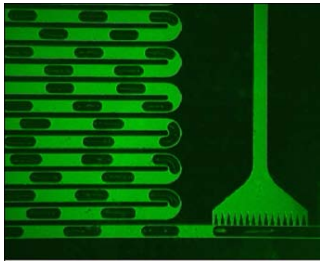
As a result as long as everyone practices the old ways and old designs and licensing of old technologies leads to profits no one wants to invest in innovation and introduction of MFS into their routine design methods.

Bench scale achieved desired conversion, yield, selectivity, productivity

Scale-up

Commercial production





Alternatives:

1. Scale-up in parallel (Scale-out, scale-up by multiplication.)

2. Scale-up vertically – account for effect of change in equipment scale on multi-scale interaction of transport and kinetic phenomena.

Typically Used Multiphase Reactor Types

- Stirred Tank (liquid, gas –liquid, liquid-solid, gas-liquid-solid)
- Bubble Column (gas-liquid, gas- (liquid –solid) (slurry)
- Packed Bed with Gas Flow (multitubular -wall cooled, adiabatic)
- Packed Bed or Structured Packing with Gas and Fine Solids Flow
- Packed Beds with Two Phase Flow (trickle beds etc.)
- Fluidized Beds (different flow regimes)
- Risers (liquid –solids, gas –solids)
- MICROREACTORS of various types.

How Can MFS Contribute?
By Embracing MFE!

SCIENCE IS ABOUT KNOWING-
ENGINEERING IS ABOUT DOING!

- Focus on a number of real multiphase systems with reaction with potential large environmental impact
- Develop on meso scale and reactor scale appropriate level models which are sufficiently generic to be utilized with different chemical systems provided physical, thermodynamic and kinetic parameters are known.
- Share the knowledge in an open and well organized form
- Use modular approach to provide codes that can be used in many industries

Methods Used in Modelling of Multiphase Flows

- DNS
- LES
- Lattice –Boltzmann
- Lagrange Euler
- **Mixture Model**
- **N- Fluid (Euler –Euler) Model**

Methods applicable to large reactor scale multiphase flow field computation need experimental validation.

Verification & Validation

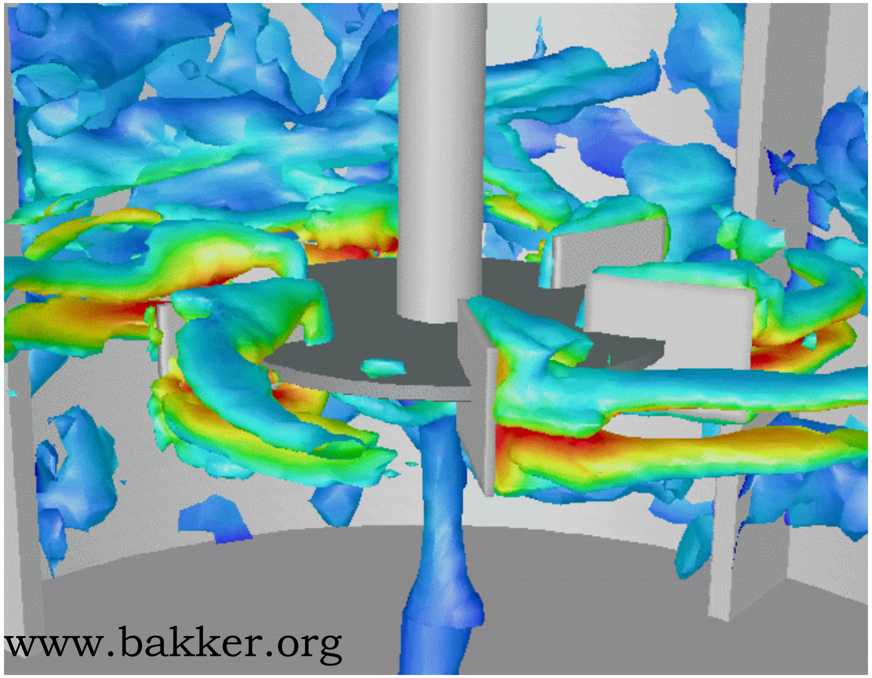
- Analyst’s paradox
  - Everyone believes an experiment except the experimentalist. No one believes an analysis except the analyst
- Verification & validation need to be given adequate attention
- Only V & V can reduce the uncertainty of CFD models and make them acceptable as ‘virtual reality’ by scientific community and regulatory authorities

Hydrodynamics and Mixing in Single and Multiphase Stirred Tank Reactors

Debangshu Guha

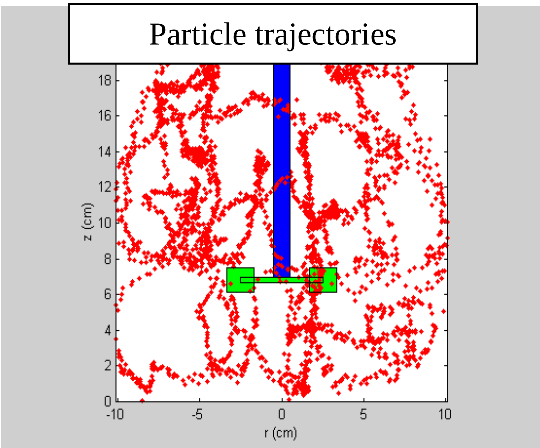
Chemical Reaction Engineering Laboratory (CREL)

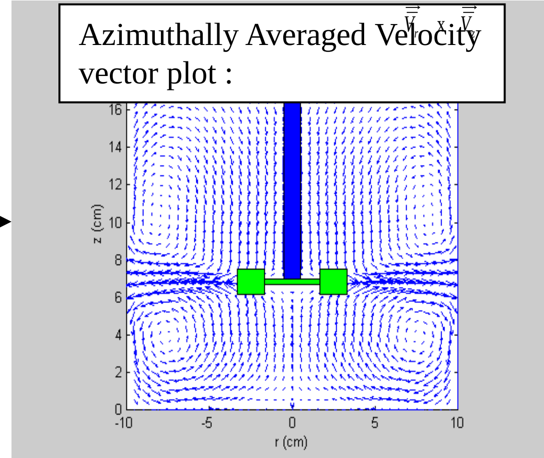
Energy, Environmental and Chemical Engineering

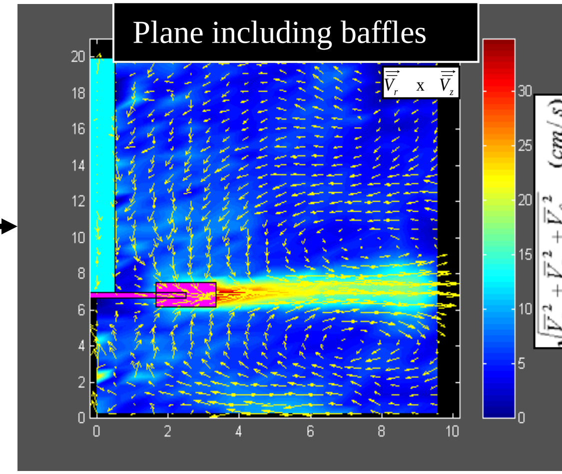


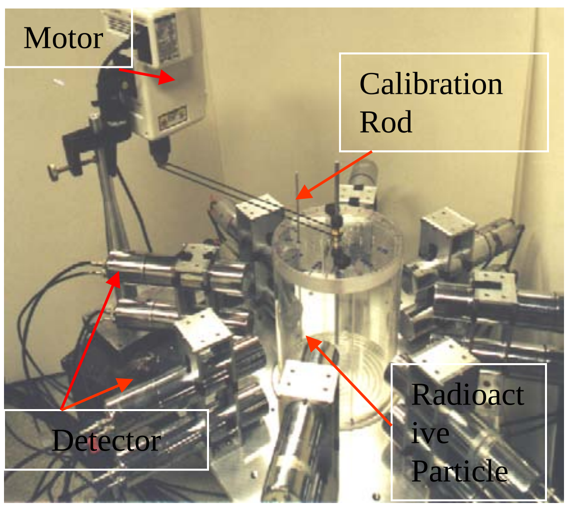
A NSF Engineering Research Center

May 22, 2007





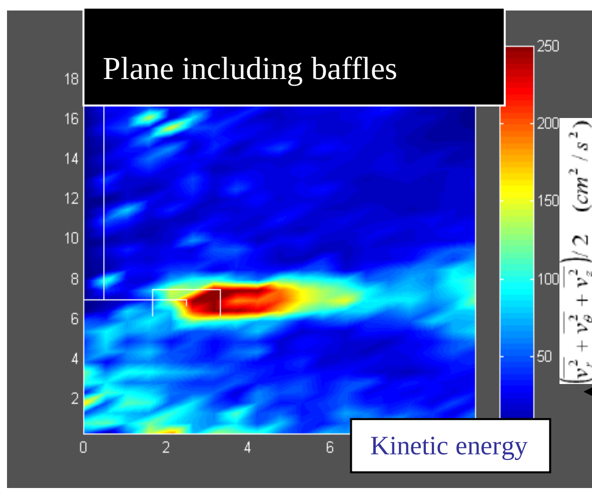


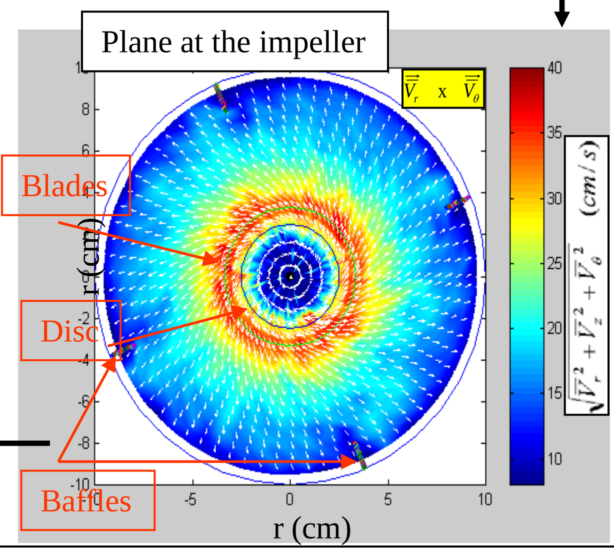


CARPT in STR :

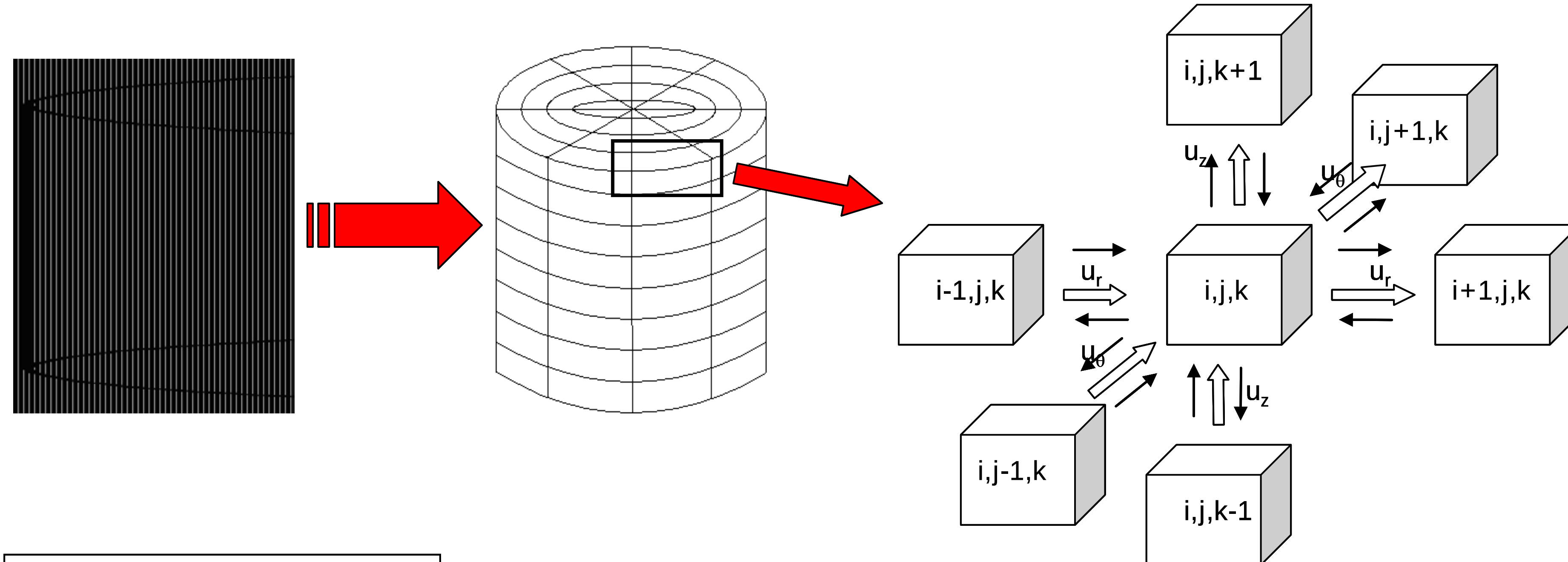
results at a glance

Rammohan et al., Chem. Eng. Research & Design (2001), 79(18), 831-844.





Overview of CFD-Based Compartmental Approach



Solve flow equations using CFD to generate the flow field in the tank

Calculate flows in and out of the compartments; Estimate the exchanges due to fluctuations

Solve **macroscopic**★ equations for all compartments simultaneously

★ Macroscopic equation consist of **convection** due to main flow, **dispersion** due to turbulence (modeled as compartmental exchange term) and the **reaction** terms

Model Equation for Single Phase System

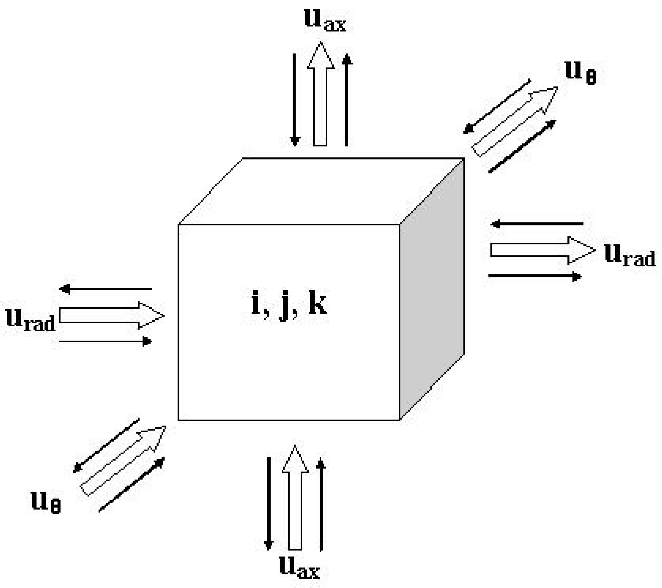
Reynolds Averaging and Volume Averaging of Continuum Species Conservation Equations → Compartment Level Equations

Turbulent Dispersion → Gradient-Diffusion Model (Boussinesq Hypothesis)

Inputs from CFD:

- ✓ Surface averaged velocity components in and out of the compartments
- ✓ Surface averaged turbulent diffusivities computed from turbulent parameters obtained by complete CFD simulation

From CFD results compartment size determined everywhere so that in each locally ***Da* ≤ 1**



Guha *et al.*, AIChE J., 2006

Mixing Effect for Multiple Reactions

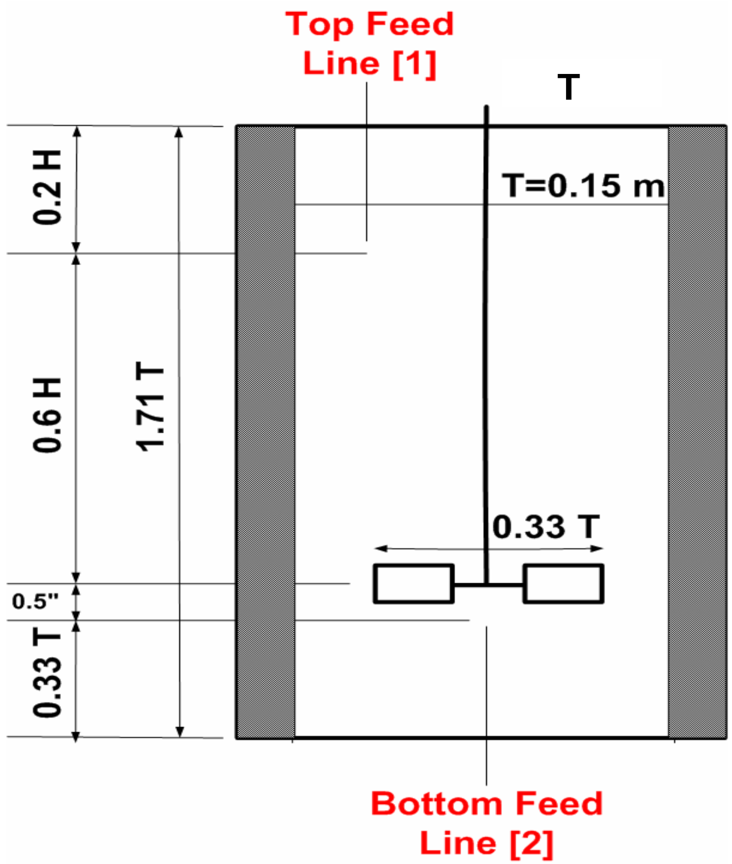
Paul & Treybal, 1971

Objective: *To illustrate the effect of feed location and mixing for a **homogeneous,** multiple-reaction system*

Reaction Scheme:

$A + B \rightarrow R$ → Desired Product

$R + B \rightarrow S$



Kinetic Constants:

$k_1 = 0.035\ m^3/mol\text{-}s$

$k_2 = 0.0038\ m^3/mol\text{-}s$

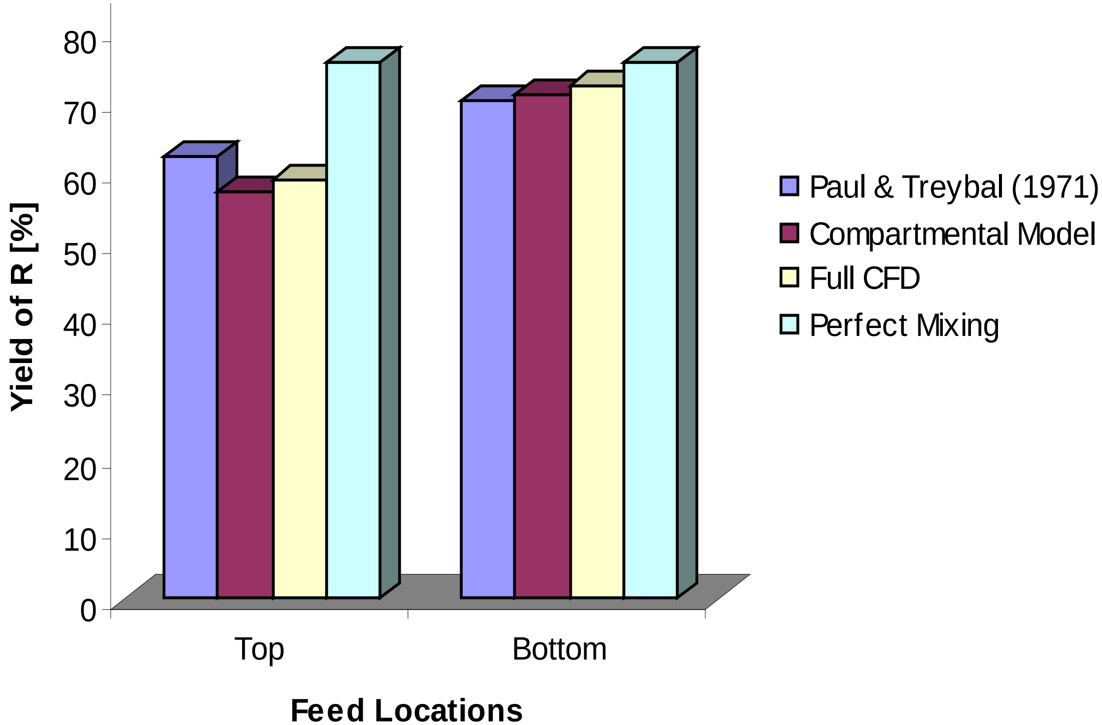
$k_1 >> k_2$

T = 0.15 m → Reactor Capacity ~ 5 litres

Yield of Desired Product

Yield of R at the completion of the reaction
Reactor Capacity: 5 liters

Guha *et al.*, AIChE J., 2006



- Impeller Speed: 1600 RPM
- Semi-batch addition of B into pre-charged A
- Initial A concentration: 200 mol/m^3
- B concentration in feed: 2000 mol/m^3
- Feeding time of B: 15 s
- Molar ratio of A to total B fed: 1:1
- Number of Compartments used: 1560 (*rxθxz:10x12x13)*
- **Yield = C_R/C_{A0}**

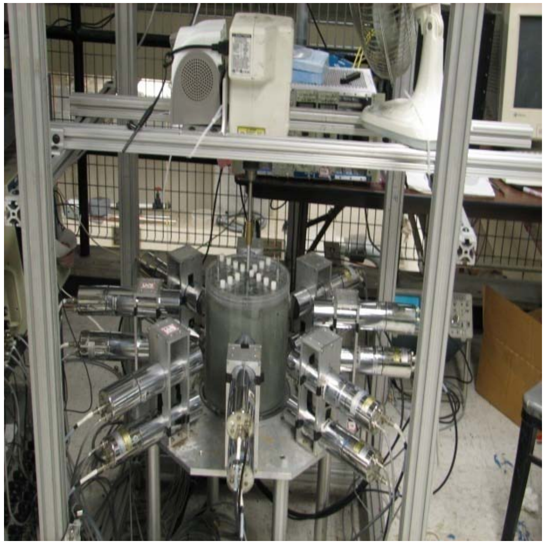
Experiment and simulation results are in reasonable agreement
Effect of feed location captured
Full CFD with species conservation did not provide much extra advantage

Experimental Conditions: CARPT/CT

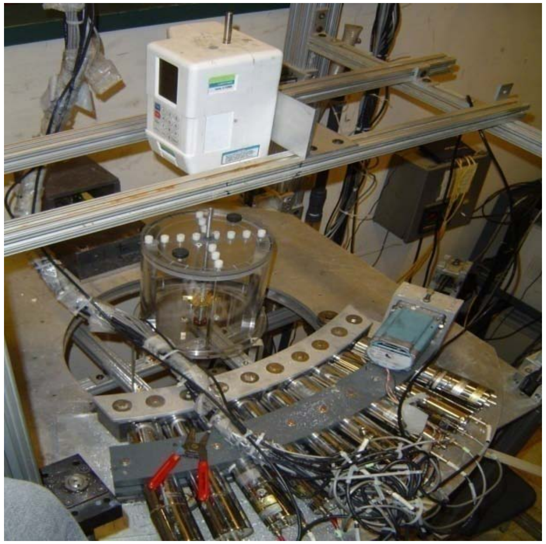
N_{js} = Just suspension speed predicted by Zwietering's correlation

Solid Hold-up (%)	N_{js} (RPM)	Expt. Set-1 (RPM)	Expt. Set-2 (RPM)
1	900	850	1000
7	1168	1050	1200

CARPT



CT



Tank Dimensions

- Diameter: 20cm
- Height: 20cm
- Impeller Diameter: 6.7cm

Solids Phase

- Material: Glass Beads
- Size: 300 microns
- Density: 2500 kg/m^3

Liquid Phase

- Material: Water

CT Scan Locations

- z/H = 0.075
- z/H = 0.25
- z/H = 0.65

Quantification of Solids Flow Field: CARPT

Distance vs. Count map from Calibration + Counts from Detectors

↓

Instantaneous Positions (x,y,z,t)

↓

Instantaneous Velocities

↓

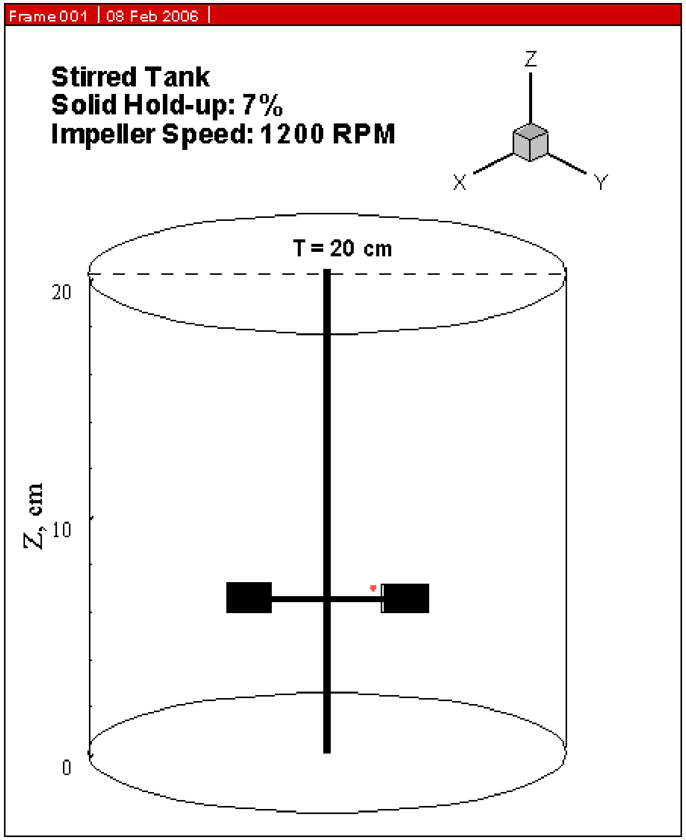
Mean Velocities

↓

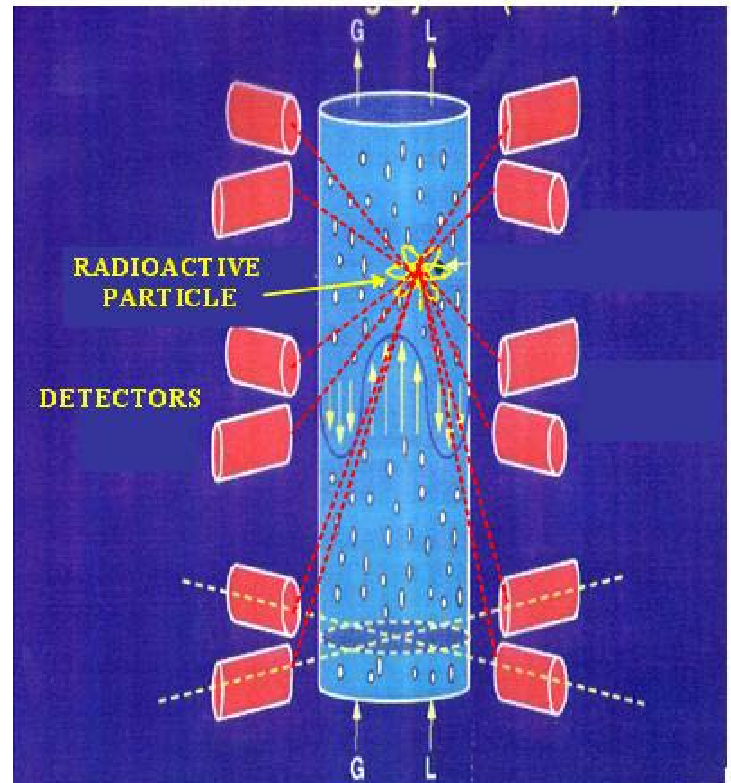
Fluctuating Velocities

↓

Turbulent Kinetic Energy



Radioactive Tracer Particle
- Same size and density as solids



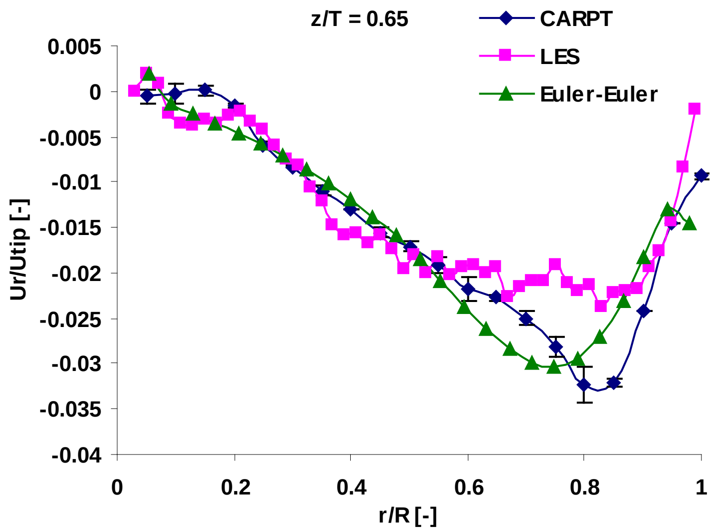
Devanathan, D.Sc. Thesis, WU, 1991

CARPT vs. CFD – Velocities and TKE (contd.)

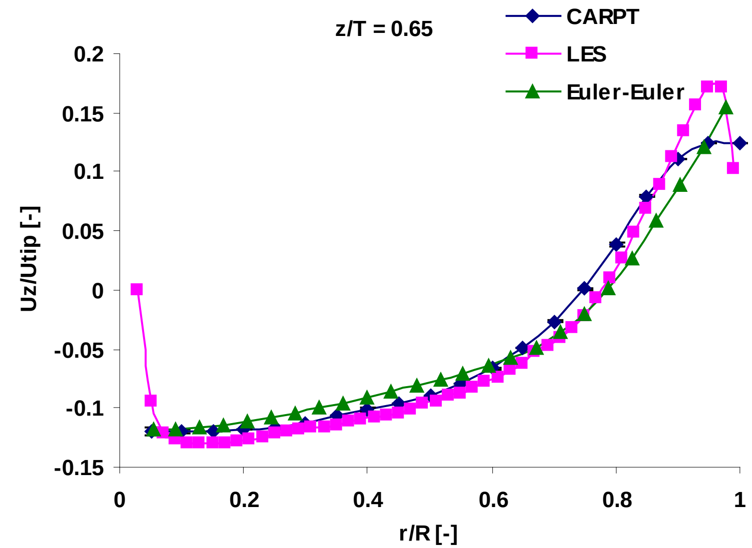
LES done by Jos Derksen, TU Delft now U. Alberta

Solids Holdup: 1% ; Above Impeller

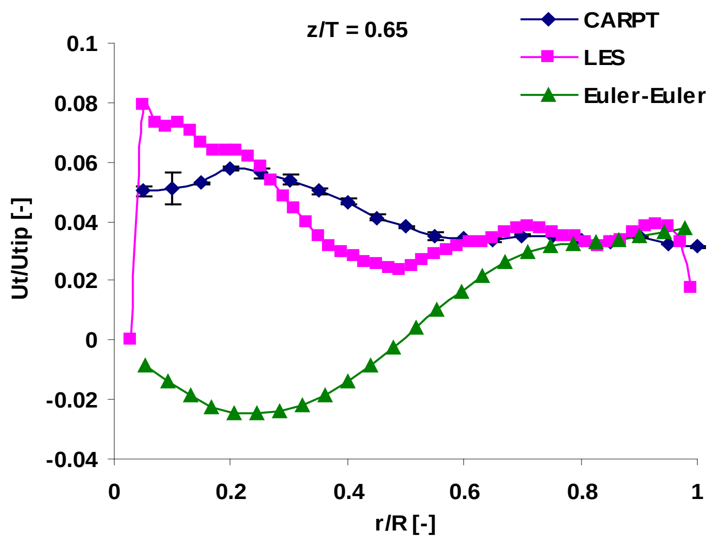
Radial Velocity



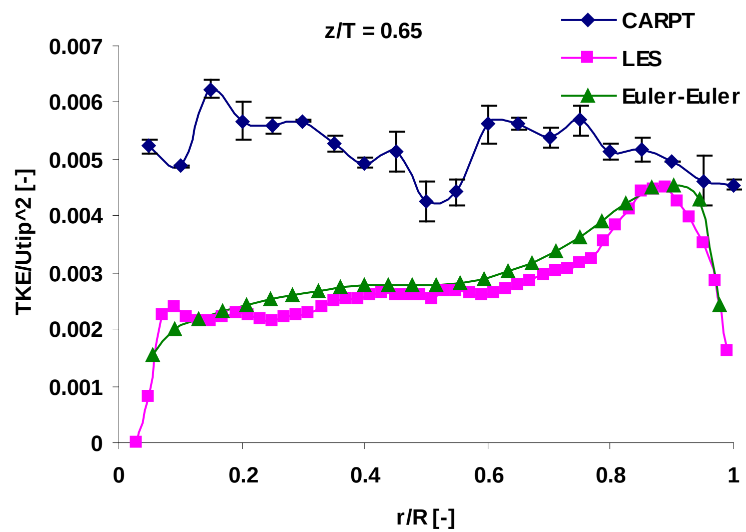
Axial Velocity



Tangential Velocity



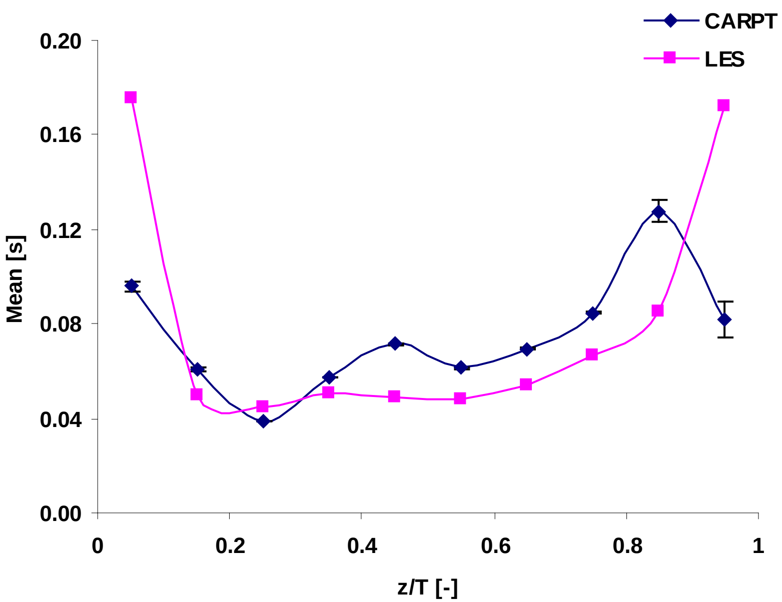
Turbulent K.E.



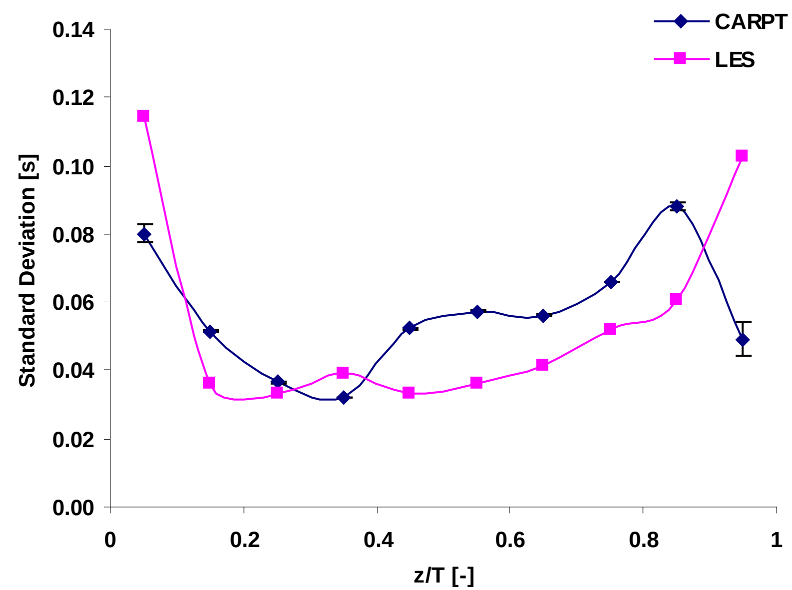
CARPT vs. CFD – Moments of STDs

Overall Solids Holdup: 1%

Mean



Standard Deviation



Reasonable predictions for moments of the STDs are obtained with LES

LES done by Jos Derksen, TU Delft now U .Alberta

Guha *et al.*, AIChE J., 2007

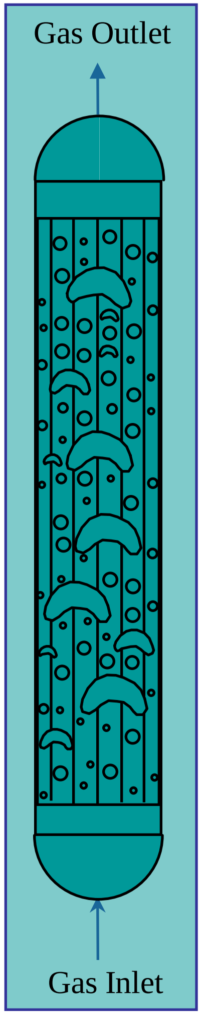
Summary and Questions

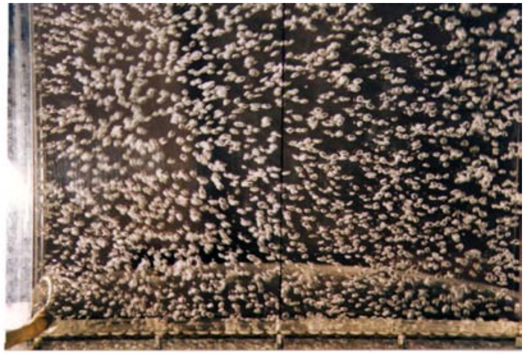
- A framework for CFD-based compartmental model developed for single phase systems
  - ✓ Determination of the number and size of compartments, extraction of flow and mixing information from CFD outlined
  - ✓ Validation with experimental data from literature for reactor performance
  - ✓ How can we do better? What level of turbulence model do we need in our calculation? How to couple kinetics with this turbulent flow? Full blown pdf approach? How to execute computation effectively?
- Solids flow dynamics obtained in a stirred tank by CARPT
  - -Solids velocities, turbulent kinetic energy and sojourn time distributions
  - -Zwietering's correlation over-predicts the impeller speed
- Neither Two Fluid Model nor LES predicts the solids dynamics revealed by CARPT or solids distribution obtained by CT
- Even mean flows are not properly predicted for gas liquid flows.
- WHAT TO DO TO IMPROVE REACTOR SCALE DESCRIPTION OF MUTIPHASE FLOWS?

BUBBLE COLUMN REACTORS

APPLICATIONS

- Fischer-Tropsch Synthesis
- Synthesis of methanol
- Coal hydrogenation
- Hydrogenation of oils
- Alkylation of methanol, benzene
- SO_2 removal from tail gas
- Effluent treatment
- Wet oxidation of effluent sludge
- Biotechnological processes
- Production of single cell protein
- Animal cell culture
- Production of biomass
- Oxidation
- Chlorination





BUBBLY FLOW

$U_G < U_{G\_T}$
- low holdup
- individual bubbles

CHURN-TURBULENT FLOW

$U_G > U_{G\_T}$
- high holdup
- large voids

Quantification of Flow Field by CARPT

Dudukovic, Oil & Gas Sci. and Tech., Rev. IFP, 55(2), 135-158, (2000)

Distance vs. Count map from Calibration
+ Counts from Detectors

Instantaneous
Positions (x,y,z,t)

Eddy
diffusivity

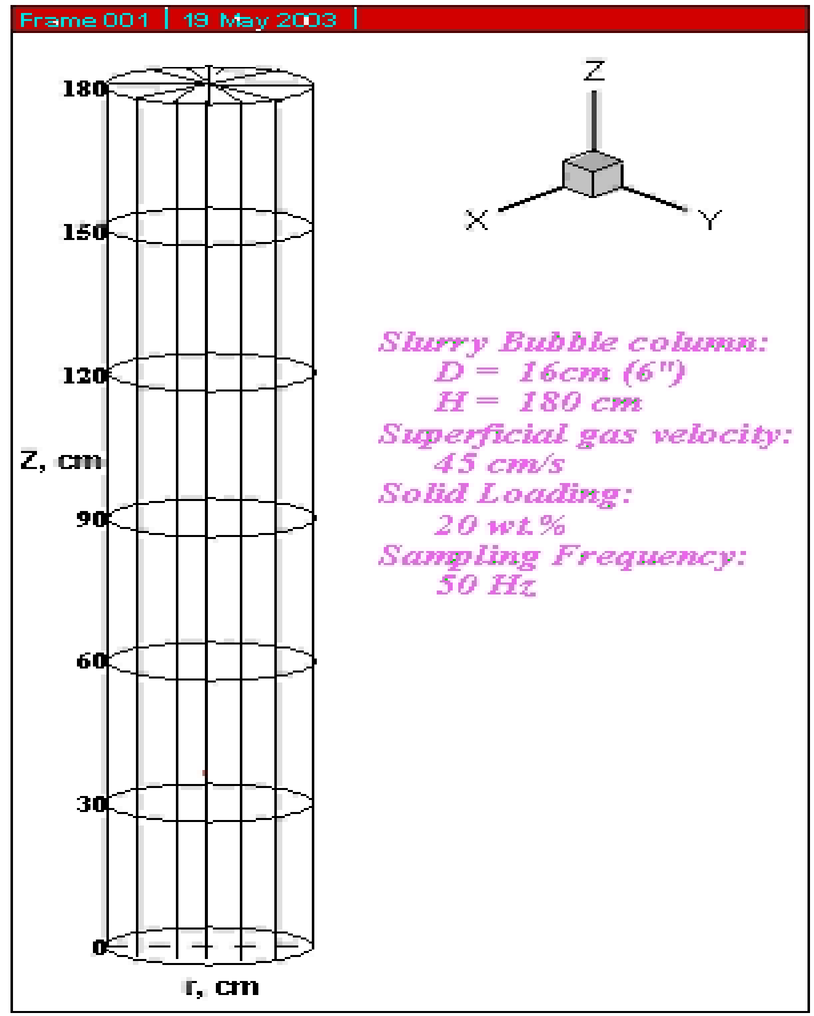
Sojourn Time
Distributions

Instantaneous
Velocities

Mean
Velocities

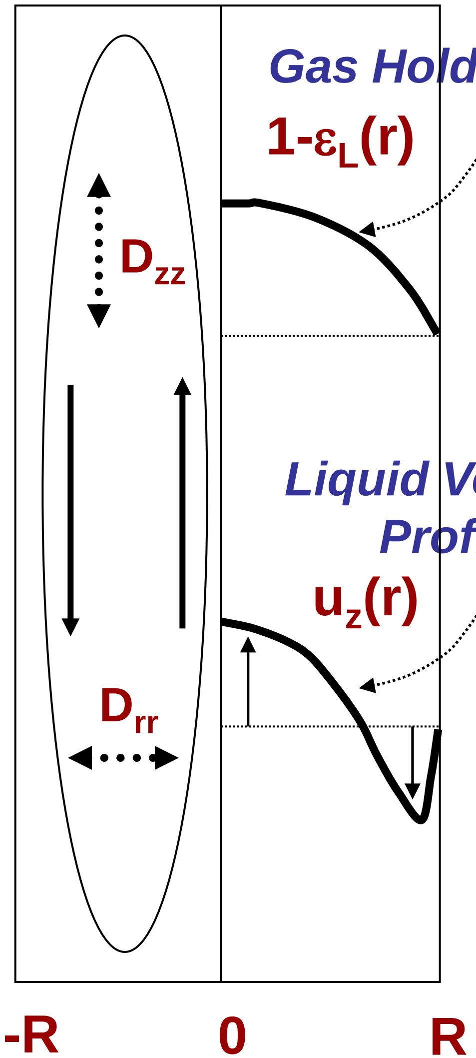
Fluctuating
Velocities

Turbulent Kinetic Energy



**Moslemian (1986);Devanathan (1990); Degaleesan (1996);
Chaouki, Larachi, Dudukovic (1997);**

CARPT-CT Experimental Input For The Time (Ensemble)-Averaged Flow and Backmixing Patterns: IMPROVED REACTOR MODEL



Transient Convection-Diffusion Equation for Liquid Mixing

$$\frac{\partial(\varepsilon_L C)}{\partial t}+\frac{\partial}{\partial z}(u_z\varepsilon_L C)=\frac{1}{r}\frac{\partial}{\partial r}\left(r\varepsilon_L D_{rr}\frac{\partial C}{\partial r}\right)+\frac{\partial}{\partial z}\left(\varepsilon_L D_{zz}\frac{\partial C}{\partial z}\right)$$

$$\overline{u_z' C'} = -D_{zr}\frac{\partial C}{\partial r} - D_{zz}\frac{\partial C}{\partial z} \quad ; \quad \overline{u_r' C'} = -D_{rr}\frac{\partial C}{\partial r} - D_{rz}\frac{\partial C}{\partial z}$$

CARPT Experiments indicate D_{zr} , D_{rz} ~ 0

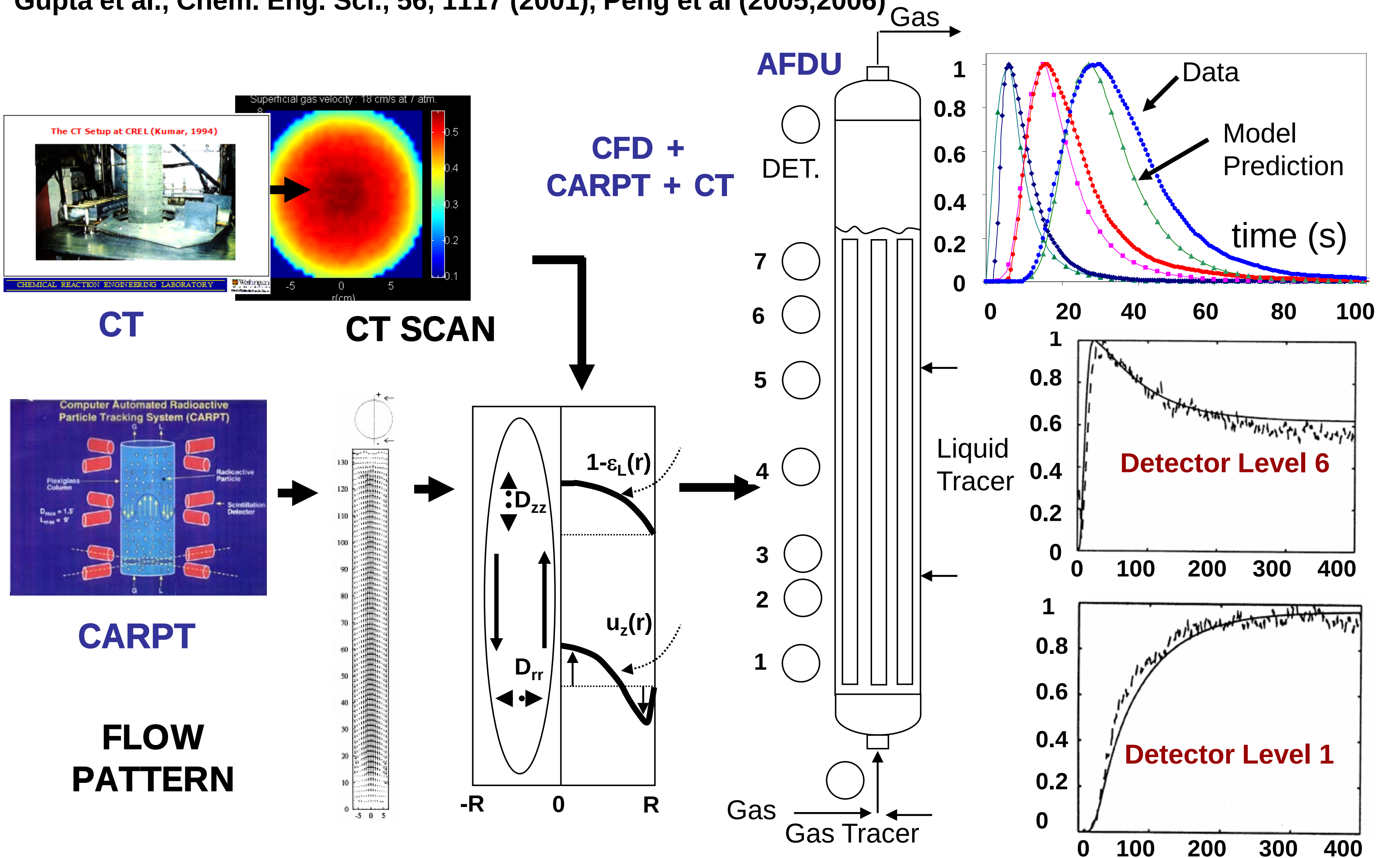
- u_z <—— **Ensemble Averaged Liquid Velocity Measured from CARPT**
- ε_L <—— **Time Averaged Liquid Holdup from CT Measurements**
- D_{zz}, D_{rr} <—— **Assumed to be CARPT Measured Diffusivities**

Bubble Column Example

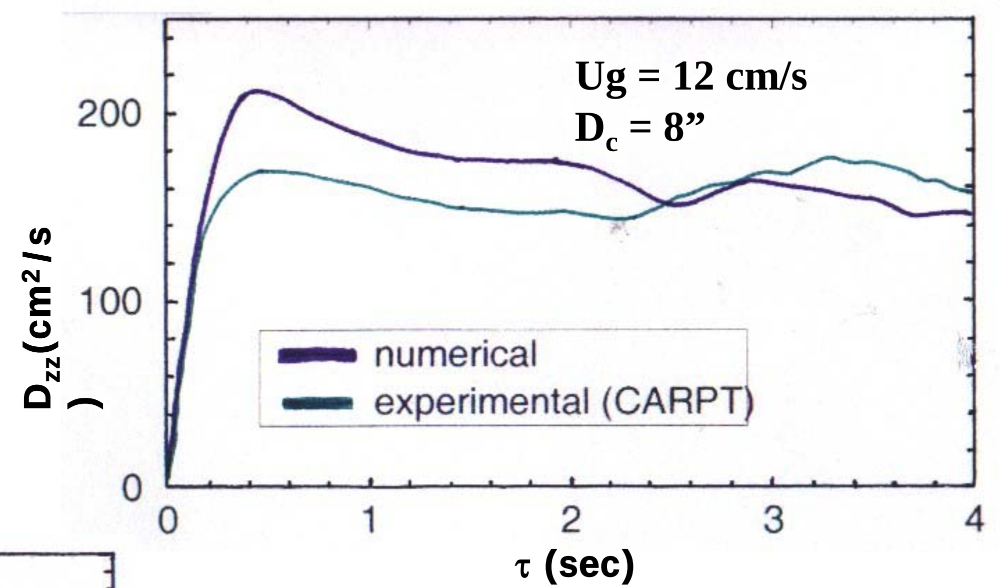
CARPT-CT and other measurements are used to develop an appropriate phenomenological reactor flow and mixing model. CFD generated data are used to assess model parameters at pilot plant or plant conditions. Reactor flow and mixing model are coupled with the kinetic information.

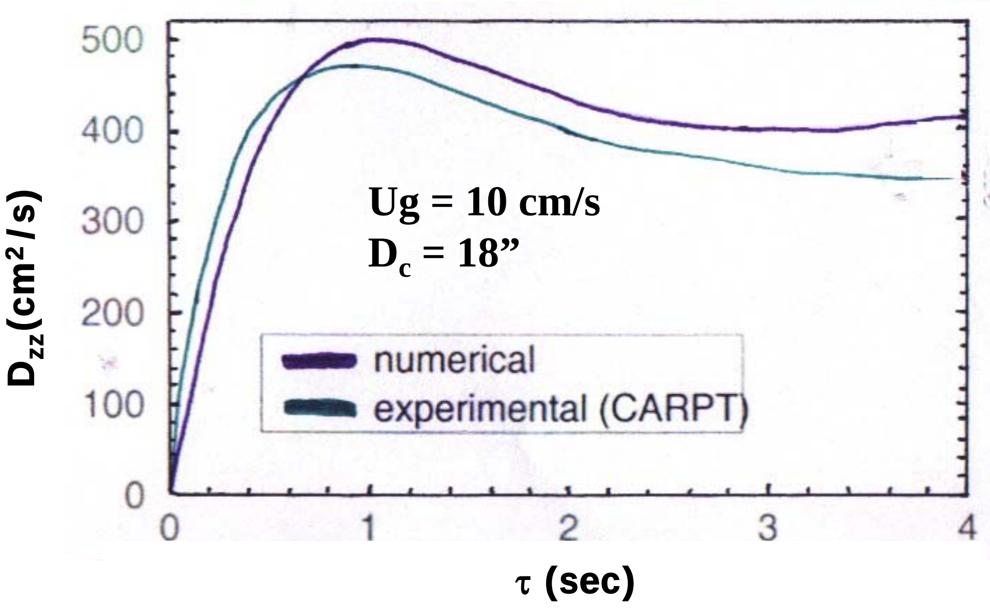
Degaleesan et al., Chem. Eng. Sci., 51, 1967(1996); I&EC Research, 36,4670 (1997);

Gupta et al., Chem. Eng. Sci., 56, 1117 (2001), Peng et al (2005,2006)



COMPARISON OF COMPUTED (CFDLIB) AND MEASURED D_{zz}





Pan et al. AICHE, 1999

Examples as to why multi-scale based scale-up and CFD should be employed to minimize the risks of commercialization of new more environmentally friendly technologies

- Solid Acid Catalyzed Alkylation

(conventional technology involves either HF or concentrated sulfuric acid as catalyst)

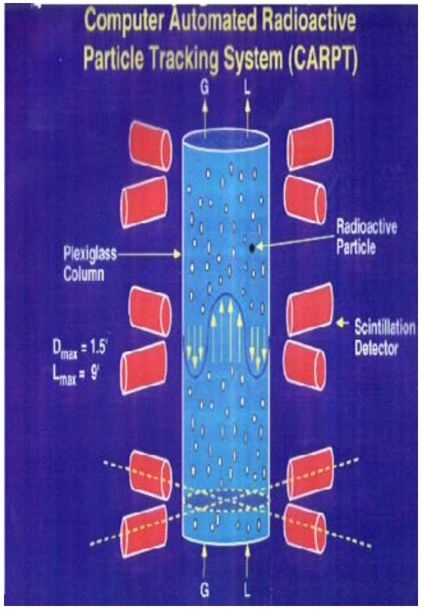
Liquid – Solid Riser

Maleic Anhydride by Partial Oxidation of Butane

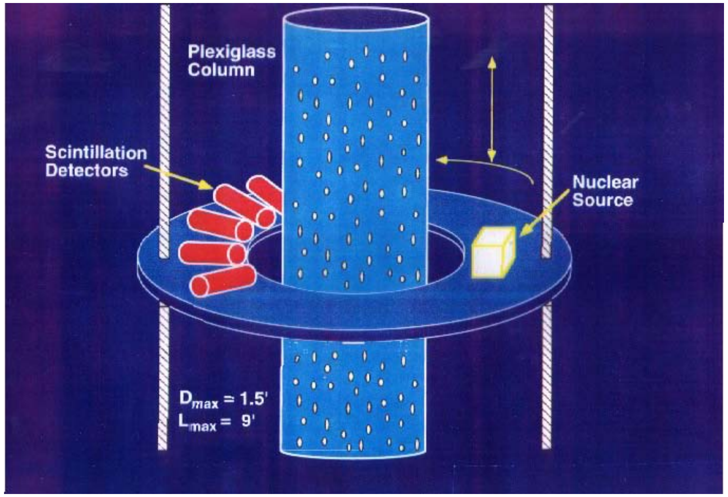
(old technology used benzene as reactant)

Gas – Solid Riser in CFB Arrangement

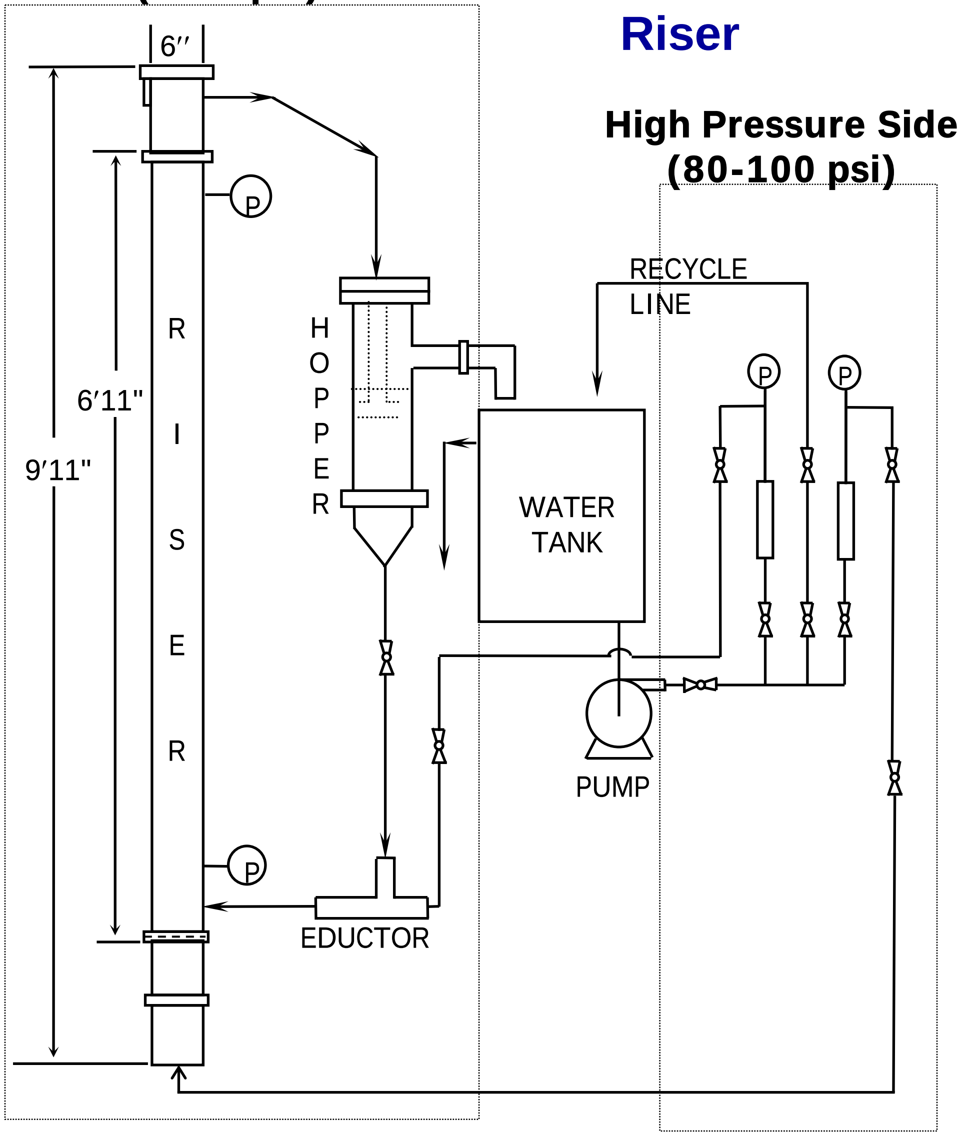
Radioactive Particle Tracking (CARPT) Provides Solids Velocity and Mixing Information



Computer Tomography (CT) Provides Solids Density Distribution



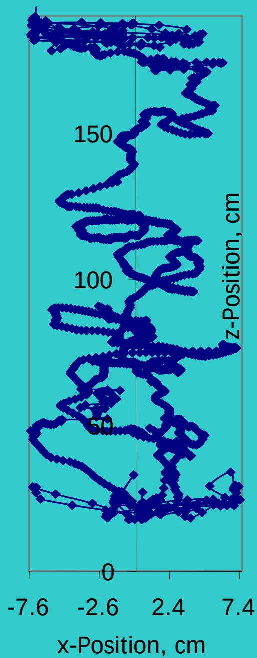
Cold Flow Model of LS Riser

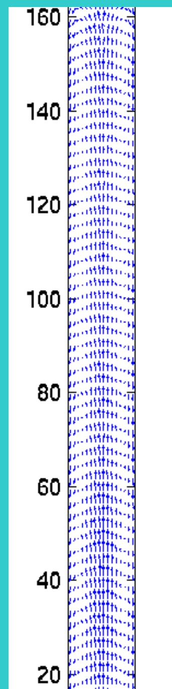


Tracer Studies Confirm Liquid In Plug Flow (N > 20)

(Devanathan, 1990; Kumar, 1994; Roy, 2000)

CARPT Results



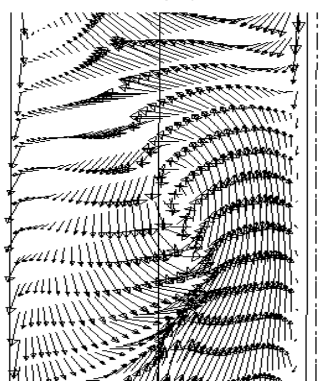


-505

CFD Results

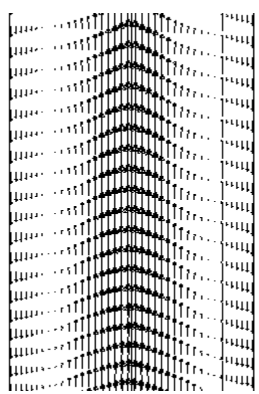
Trace over 38 s (1900 positions)

Z = 125 cm



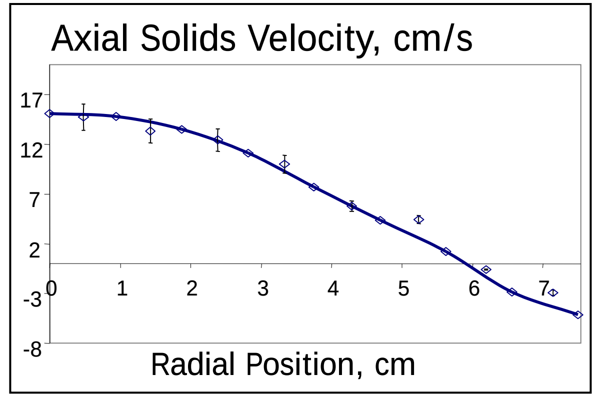
Z = 100 cm

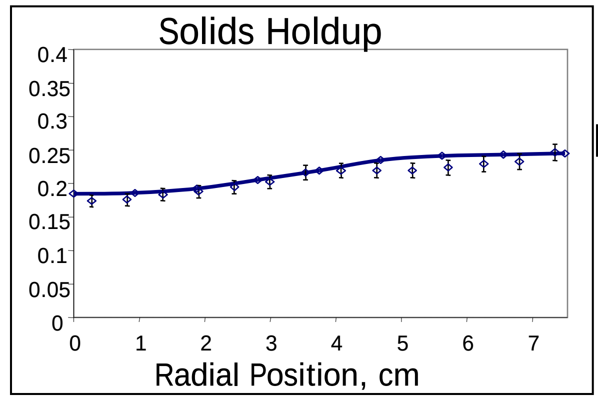
t = 60 s

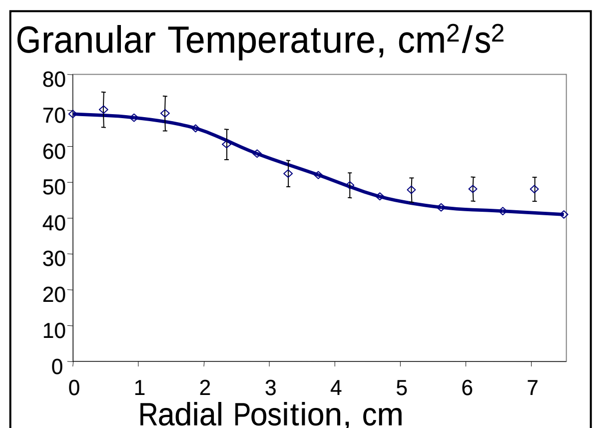


Time Average
(25 - 100 s)

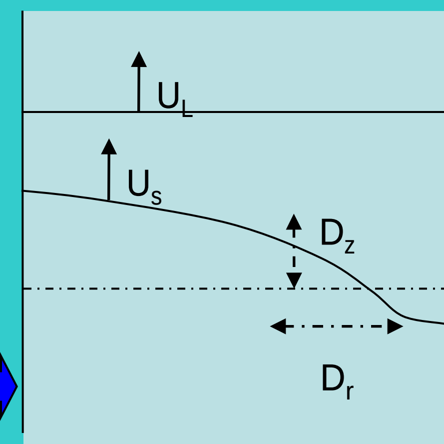
Comparison of CFD with Data







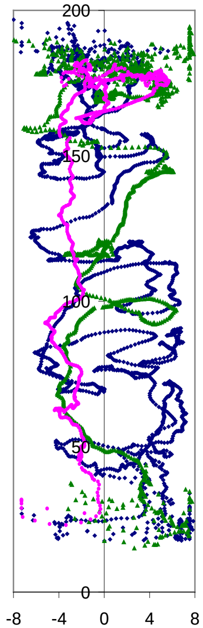
Final 2-D Convection Diffusion Reactor Model for the Riser



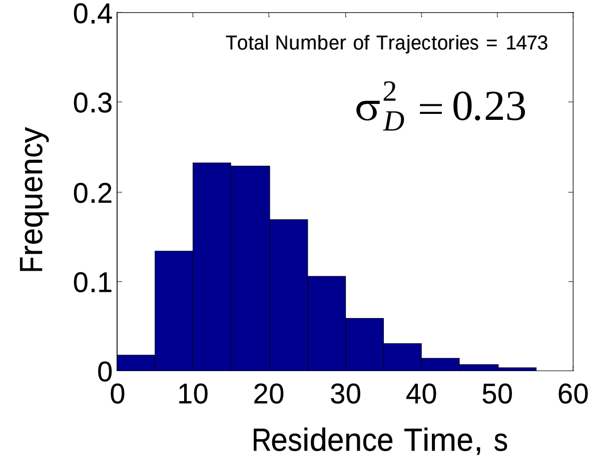
Ready for plant design, optimization and model based control.

Roy et. al, 2000, 2001

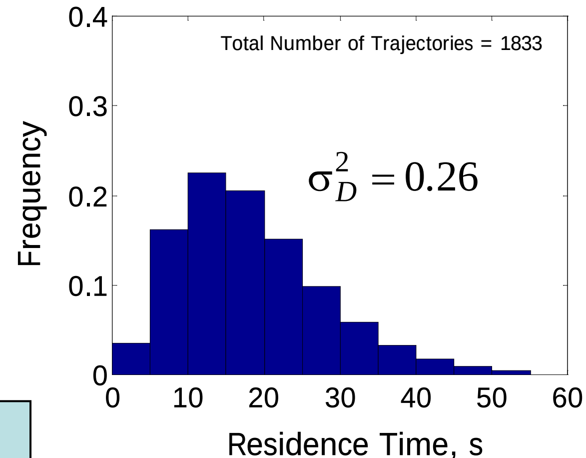
SOLIDS RESIDENCE TIME DISTRIBUTIONS



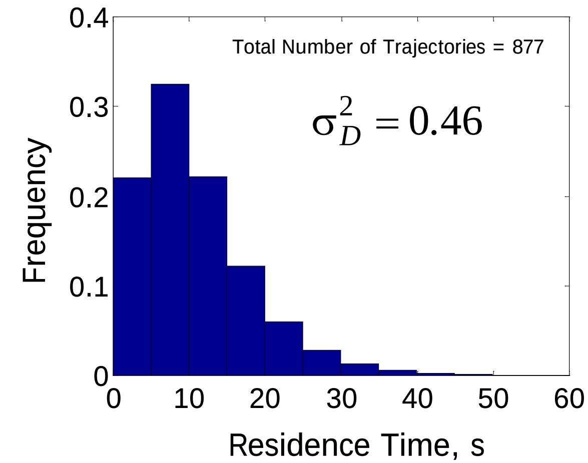
<u>Trajectories</u>



<u>U_l= 15 cm/s; S/L = 0.15</u>



<u>U_l= 20 cm/s; S/L = 0.10</u>



<u>U_l= 23 cm/s; S/L = 0.20</u>

$2 \leq N_{solids} < 6$

OVERALL $0.18 \leq \sigma_D^2 \leq 0.61$

Concept of Atom and Mass Economy

Atom economy is a measure of how efficiently raw materials are used. (Benzene route 18/42=.43; n-butane route 9/17=0.53)

Example: Mass economy of Maleic anhydride production via benzene & n-butane route.

Benzene route:

$$\boxed{2C_6H_6 + 9O_2 \xrightarrow{V_2O_5\ MoO_3} 2C_4H_2O_3 + H_2O + 4CO_2}$$

$$Mass\ Efficiency = \frac{2(4)(12) + 3(2)(16) + 2(2)(1)}{2(6)(12) + 9(12)(16) + 2(6)(1)} \times 100 = 44.4\%$$

n-Butane route:

$$\boxed{C_4H_{10} + 3.5O_2 \xrightarrow{(VO)_5\ P_2O_5} C_4H_2O_3 + 4H_2O}$$

$$Mass\ Efficiency = \frac{(4)(12) + (3)(16) + (2)(1)}{4(12) + 3.5(2)(16) + 10(1)} \times 100 = 57.6\%$$

Partial Oxidation or Butane to Maleic Anhydride (Replaced Benzene Oxidation)

$$C_4H_{10} + \frac{7}{2}\ O_2 \rightarrow C_4H_2O_3 + 4\ H_2O$$

$$C_4H_2O_3 + O_2 \rightarrow 4\ CO + H_2O$$

Vanadium Pentoxide Catalyst

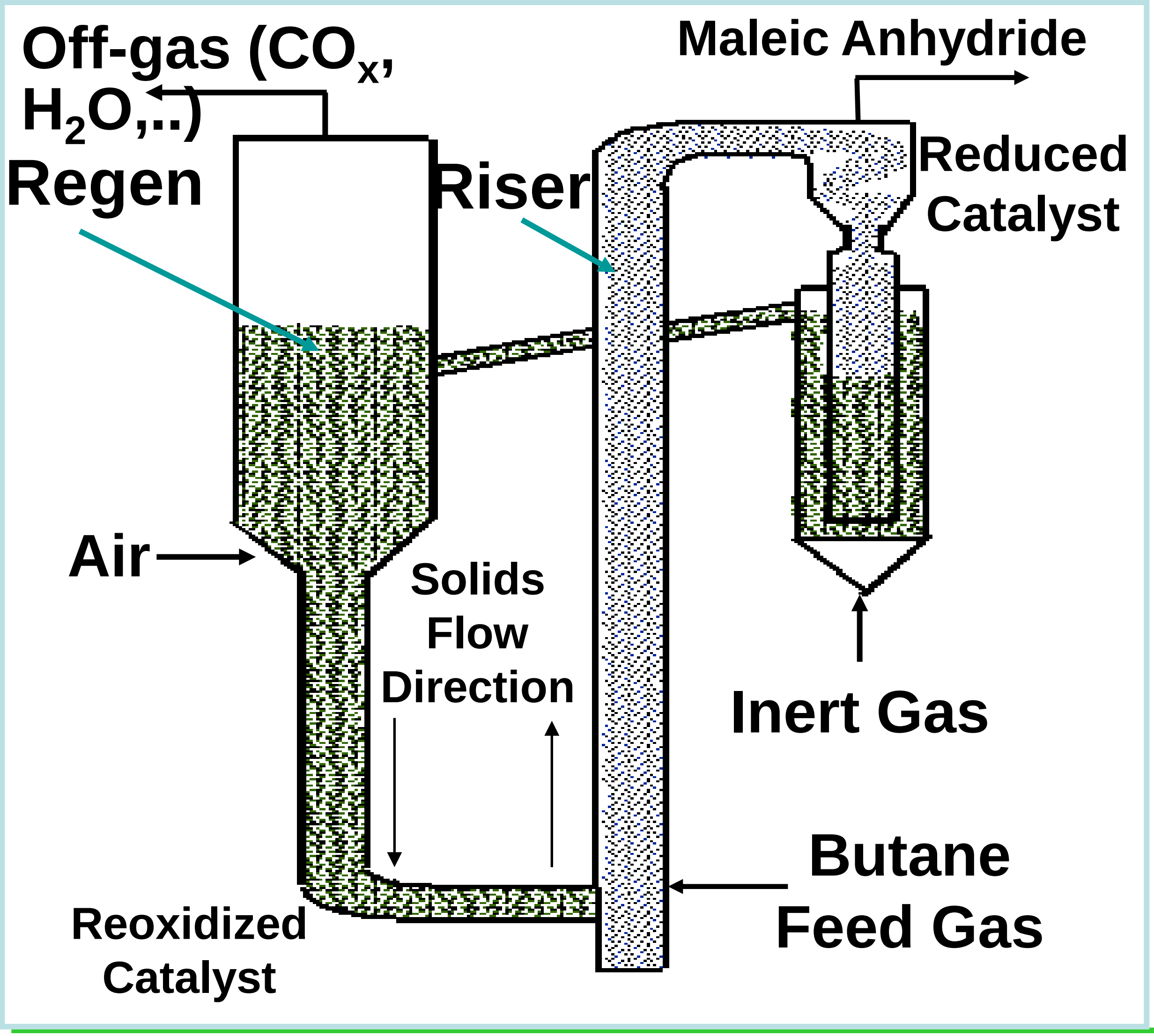
$$C_4H_{10} + \frac{11}{2} O_2 \rightarrow 2\ CO + 2\ CO_2 + 5\ H_2O$$

All reactions are exothermic and the heat is removed using cooling of reactor wall (tubular reactor) or via heat exchange pipes (fluidized bed reactor). The amount of butane in the feed (1.8% for packed beds and 4% fluidized beds) is controlled as not to form an explosive mixture. Hence, low concentration of butane results in low yield of maleic anhydride (1% in product) which requires costly separation of product mixture.

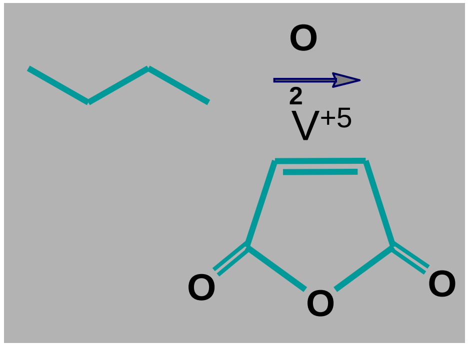
Industrial reactors:

1. Packed beds
2. Fluidized beds (ALMA Process)
3. CFB reactor (DuPont Process)

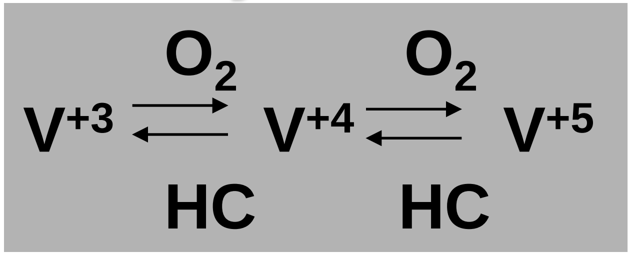
Circulating Fluid Bed (CFB) Reactor for Butane Oxidation



Main Reaction

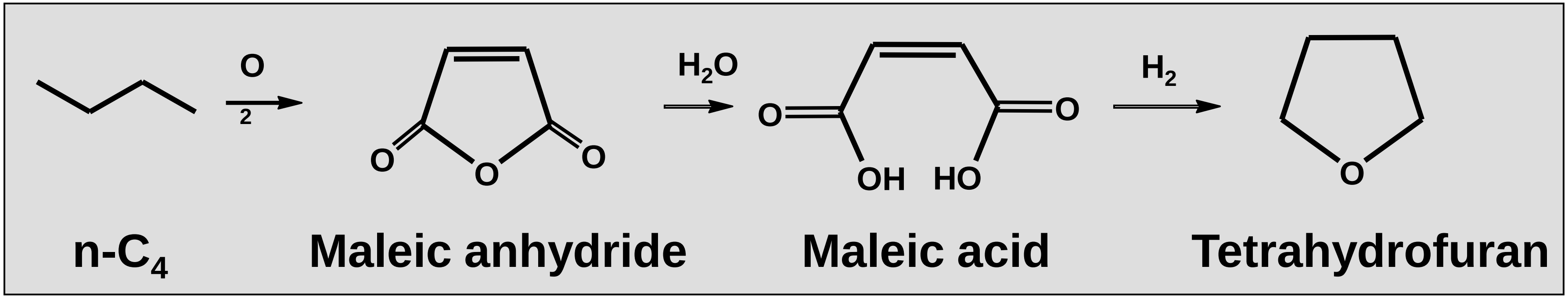


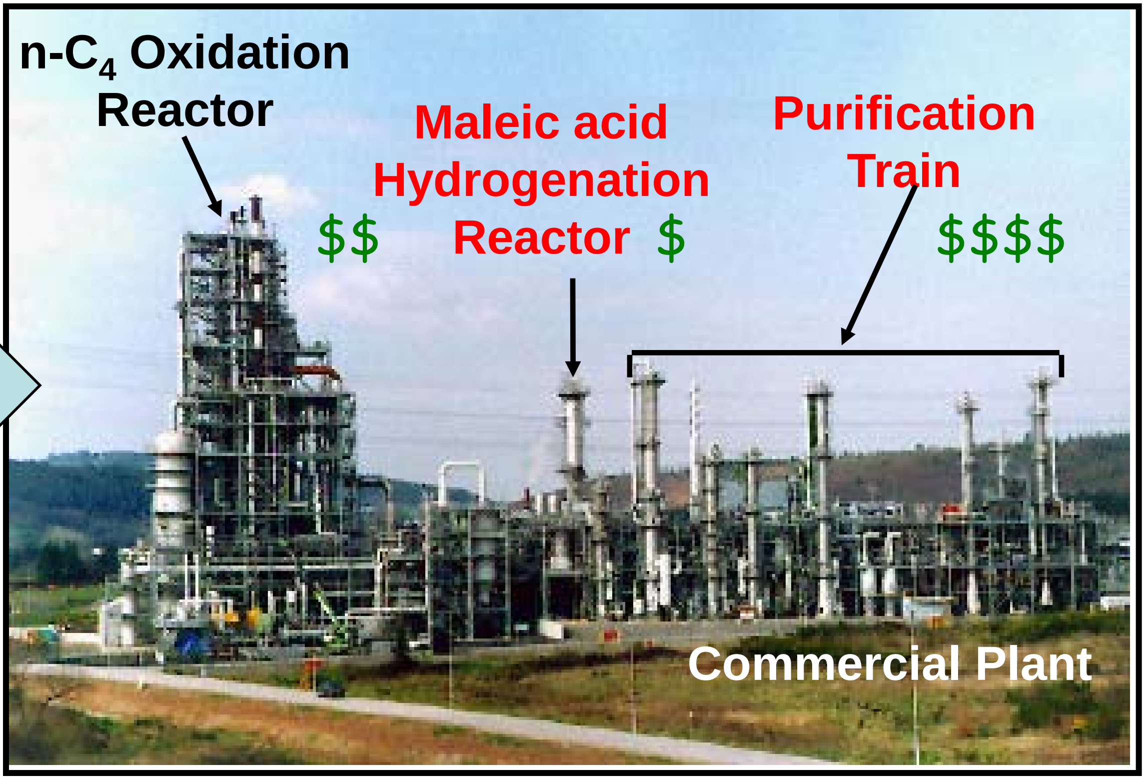
Catalyst Redox



Riser

Example: Butane to THF Process



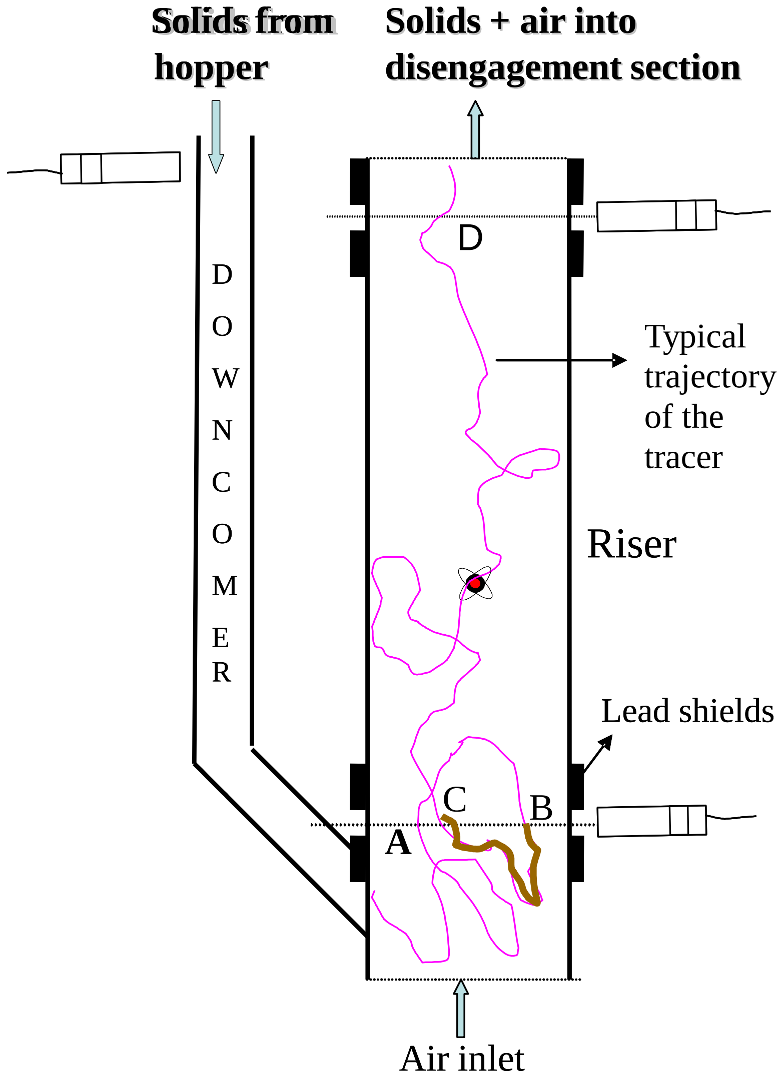


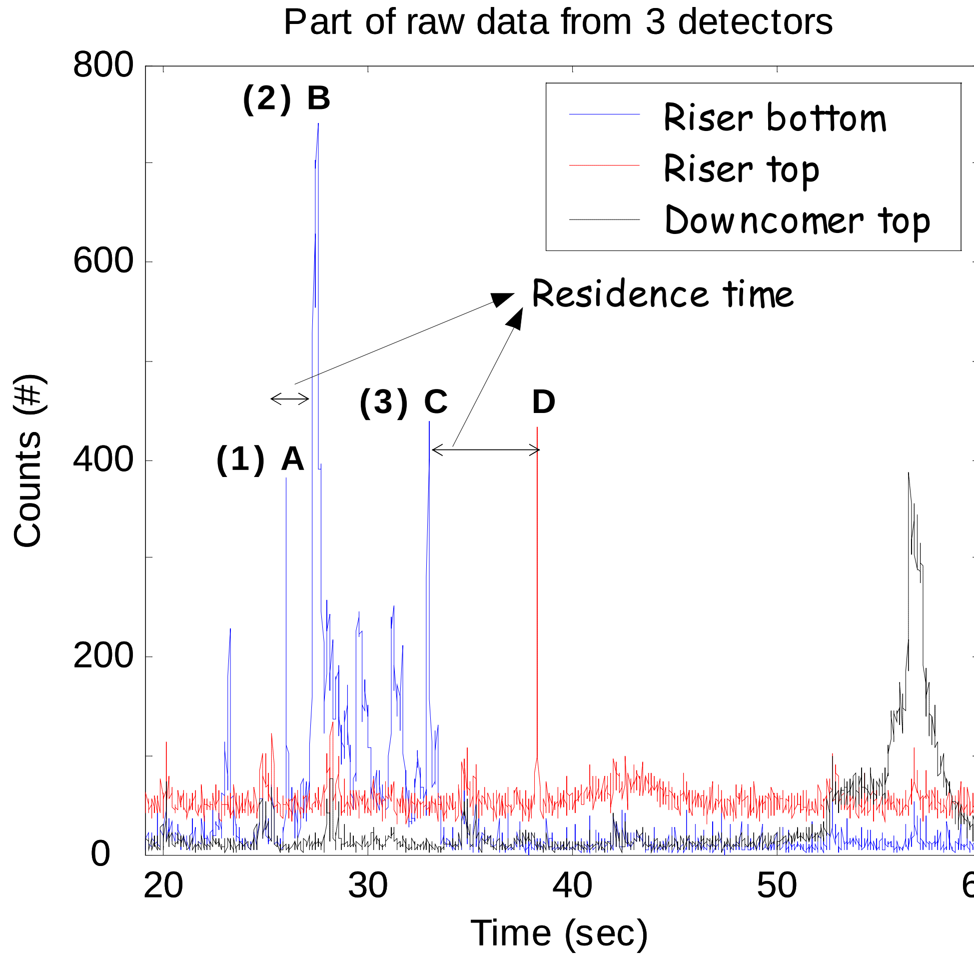
Scale-up of CFBs requires at the minimum matching the mean and variance of contact times in the riser and in the fluidized bed for the pilot and plant scale

- This is hard to do when solids holdup in the two vessels is not precisely known and when solids circulation rate is unknown

- CT and CARPT can determine this and provide a data base for CFD validation in prediction of these important parameters

Evaluation of Residence Time and First Passage Time Distributions from CARPT Experiments: Part of MFDRC Initiative

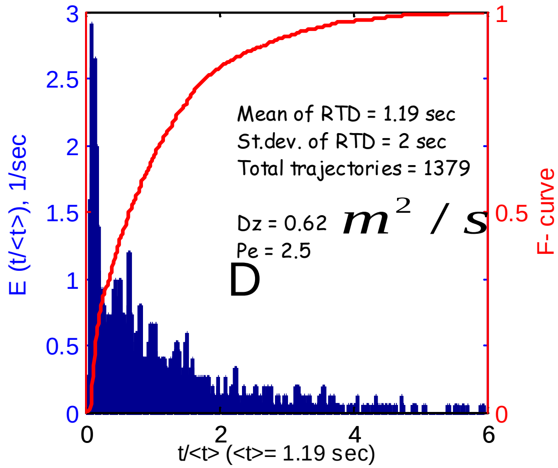


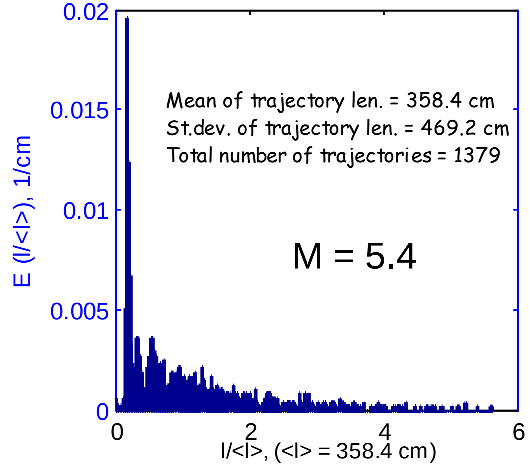


Time spent by the tracer between B-C should not be counted in the residence time

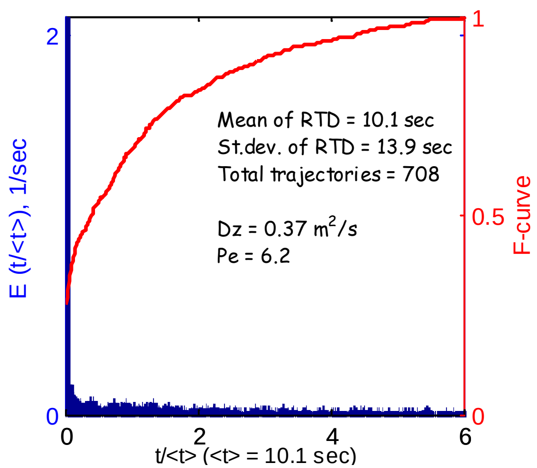
Solids Backmixing – RTD, TLDs

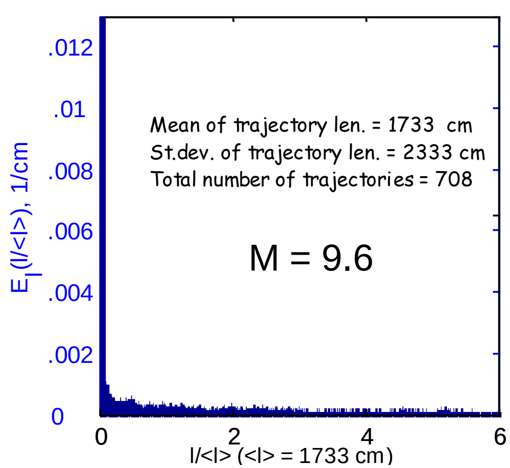
FF - U_g^{riser} = 3.2 m.s^{-1}; Gs = 26.6 kg.m^{-2}.s^{-1}





DPT - U_g^{riser} = 5.49 m.s^{-1}; Gs = 102 kg.m^{-2}.s^{-1}





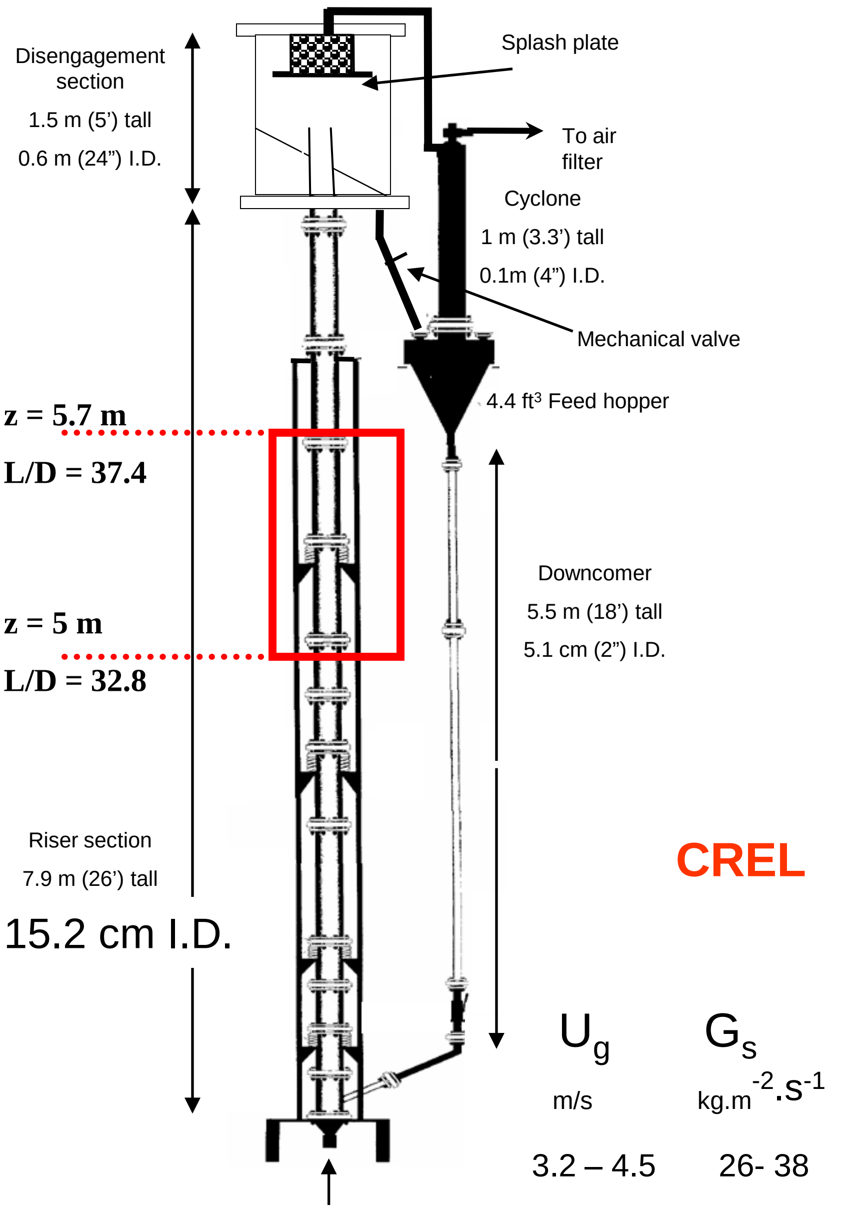
Bhusarapu et al., 2005, *I&ECR*.

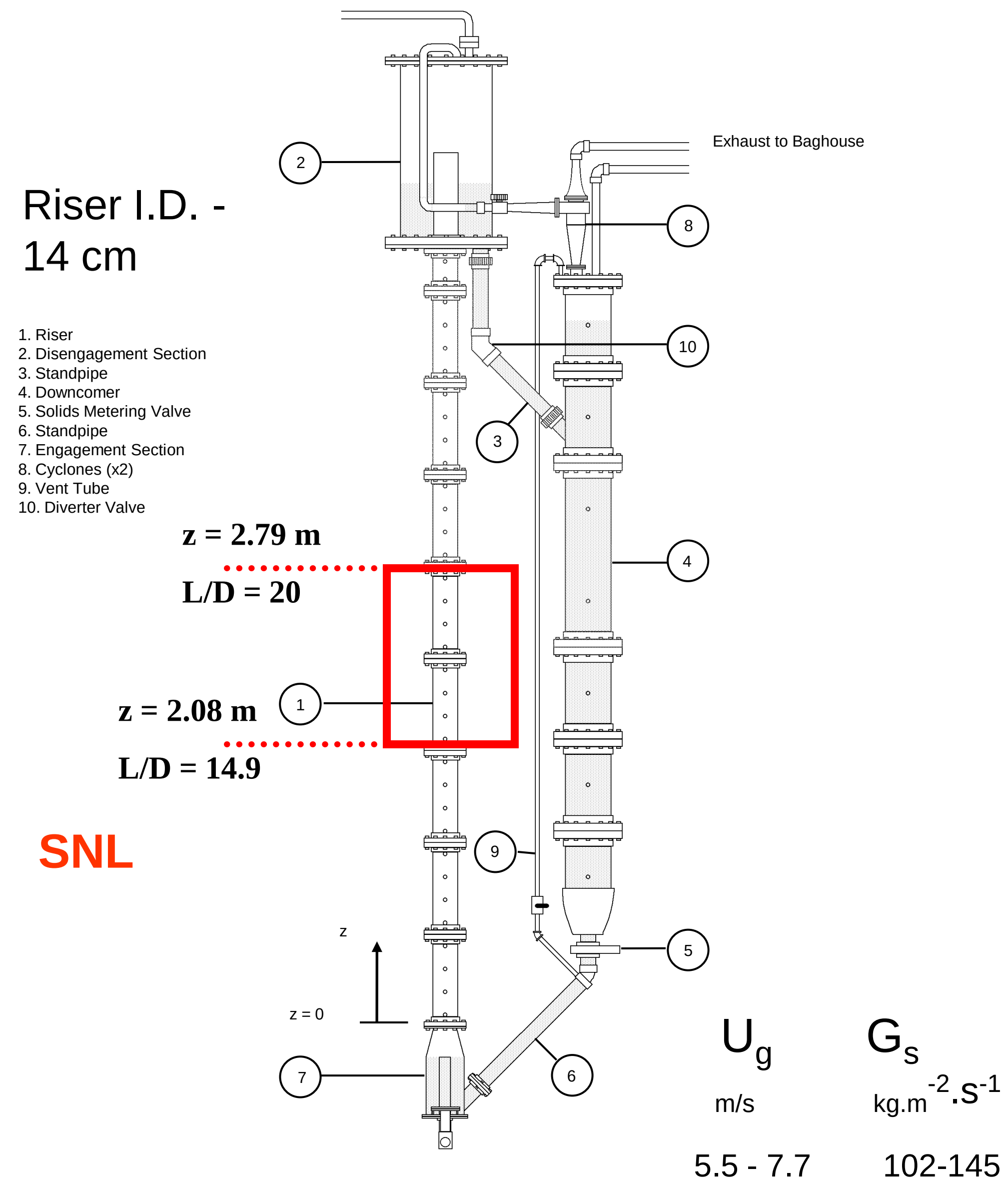
MFDRC Initiative

- Core-annulus flow structure in riser results in a RTD with extended tail in the DPT regime, while in the FF regime it results in a hint of a dual peak along with the extended tail.
- Axial dispersion increases with solids mass flux at fixed gas velocity (not shown)
- 'Macromixing index' decreases with flux at fixed gas velocity (not shown)

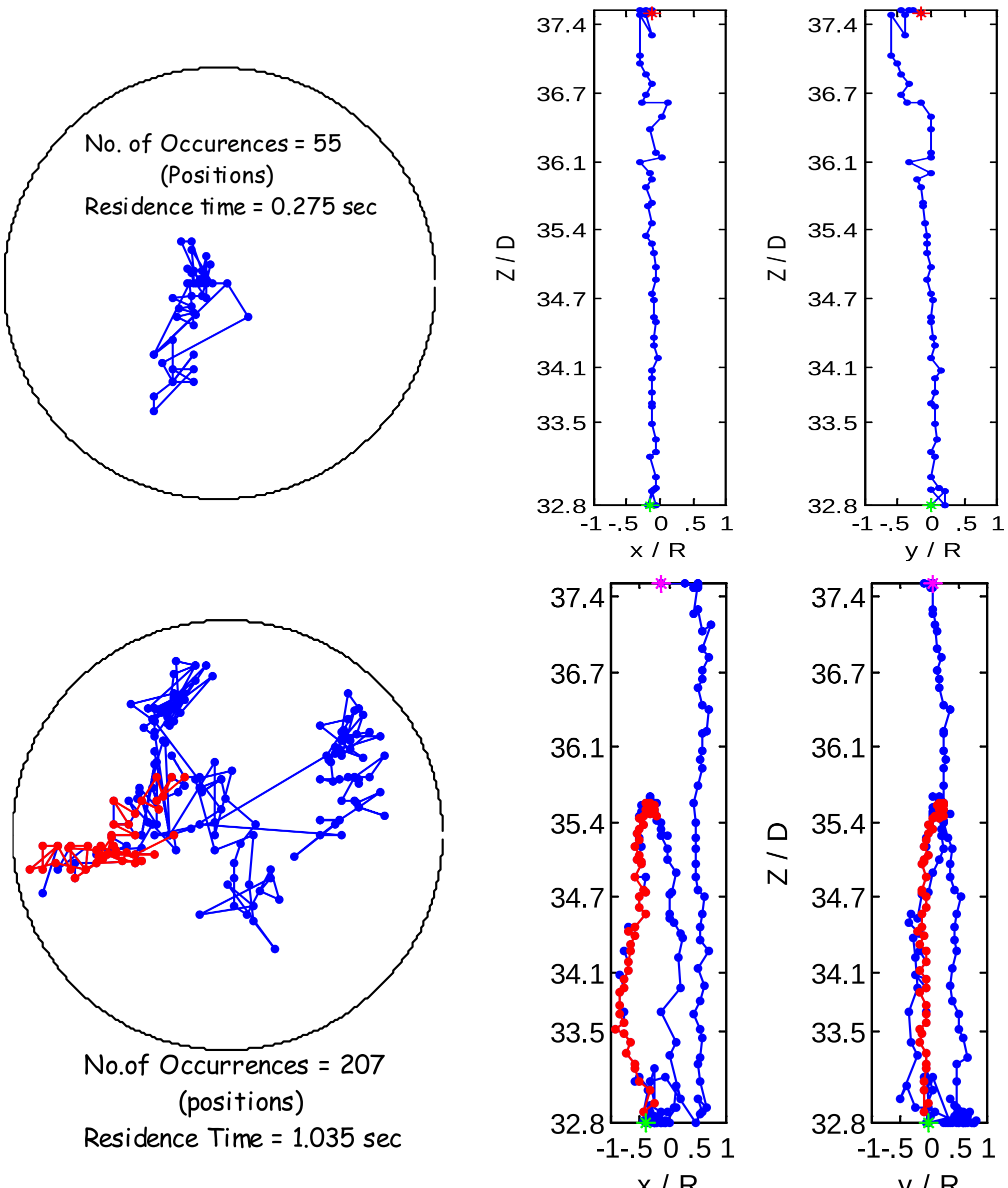
$$M = \frac{Mean \cdot of \cdot trajectory \cdot length}{Characteristic \cdot length}$$

Experimental Setup – CFBs





Instantaneous Particle Traces – FF Regime (MFDRC)



U_g^{riser} = 3.2 m.s^{-1}

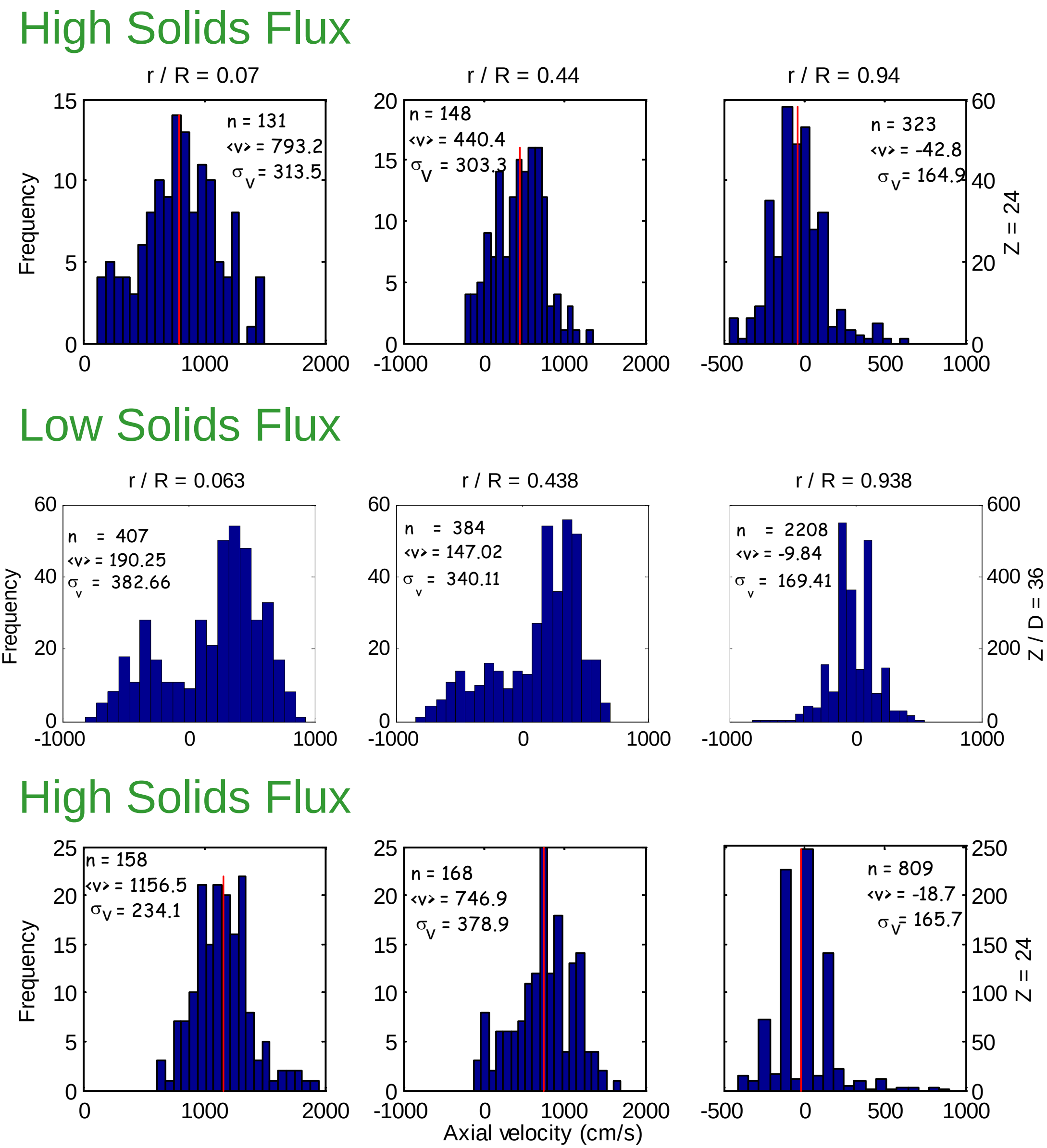
G_s = 26.6 kg.m^{-2}.s^{-1}

Zone of Investigation (Z/D) – 33.5-36.7

- Few times tracer passed through the section straight, while many more times tracer underwent internal recirculation in the section.
- Tracer downflow (negative axial velocity) near the center(core region) observed.
- Span of residence times – 0.1-100 sec! Three orders of magnitude !!

Bhusarapu *et al.*, 2005a, *Powder Tech.*,

High and Low fluxes – Axial Velocity PDF's (MFDRC)



n-Number of occurrences in the voxel (#); <v>-Mean axial velocity (cm/s); σ_V -Standard deviation of the velocity (cm/s)

FF Regime –

- Bimodal in low, uni-modal at high fluxes- center
- No negative vel's at center at high fluxes

→ Clustering phenomenon prevalent across CS – low

→ Mainly near walls - high

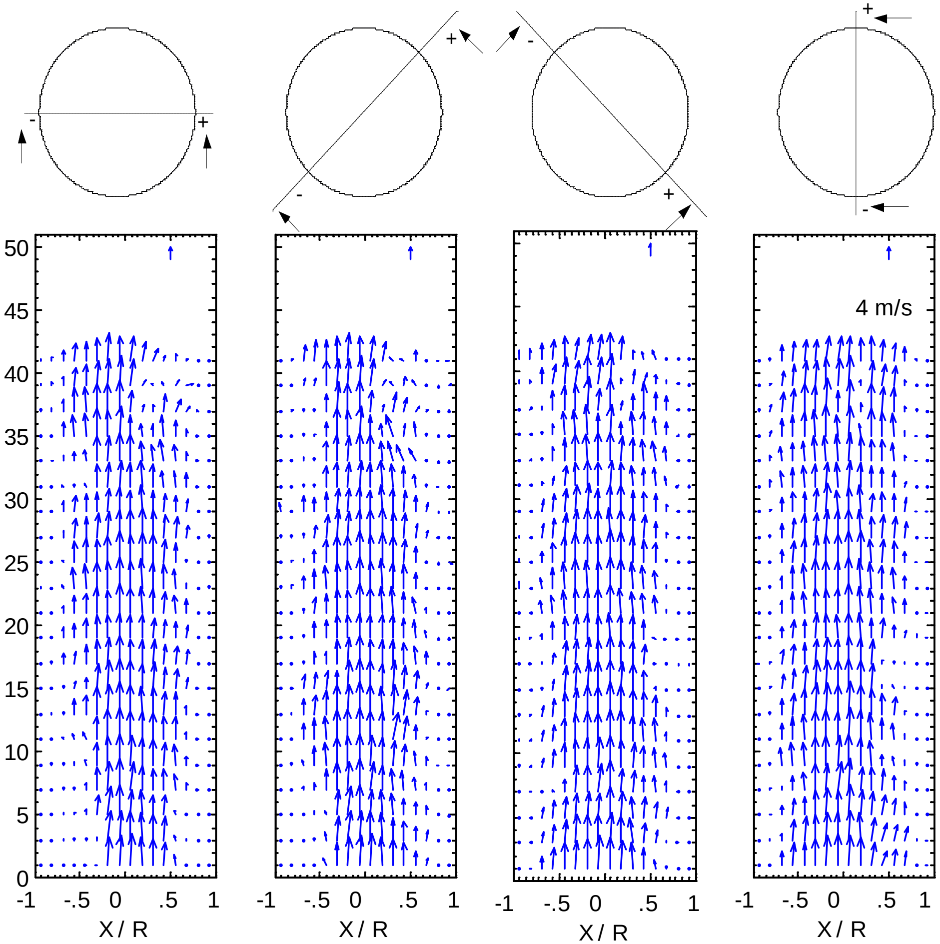
DPT Regime –

- Time-averaged vel's are negative near the wall - high

→ Downflow velocity in the annulus increases with flux

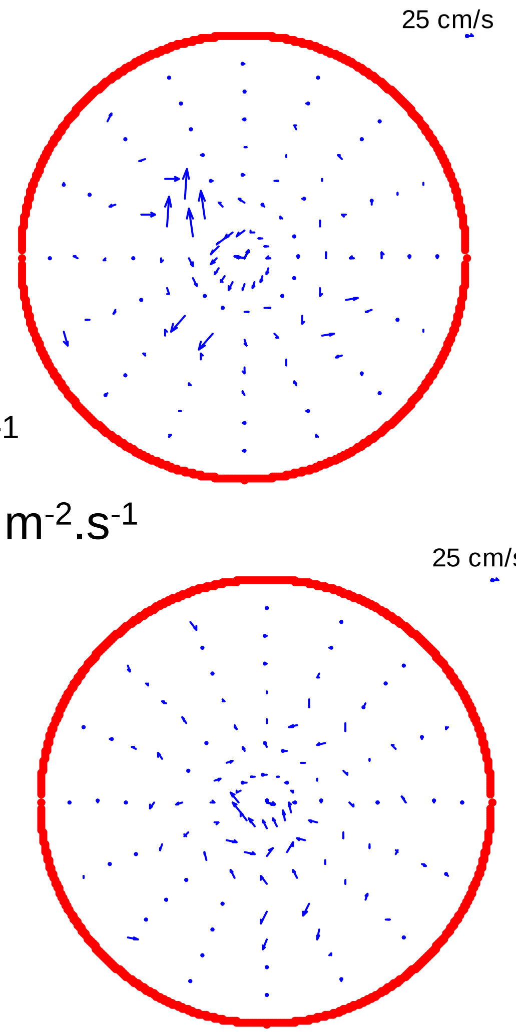
Mean Velocity Field – High Flux (FF Regime) – SNL (MFDRC)

Bhusarapu et al., 2005b, *I&ECR*, In review



Ug = 5.56 m.s^{-1}

Gs = 144.5 kg.m^{-2}.s^{-1}



- Axi-symmetric with negligible radial and azimuthal velocities
- Little axial variation – “fully-developed”
- Strong core-annular structure; Similar observations in DPT regime

J.A.M. (Hans) Kuipers et al. DENSE GAS-SOLID FLOWS & MODELLING

GAS-PARTICLE SYSTEMS

+ very broad range of applications and related equipment geometries
+ occurence of both dilute and dense particle-laden flows (poly-disperse)
+ display a great variety of (very complex) flow structures
+ flows are inherently unsteady (bubbles, clusters)

IMPLICATIONS FOR MODELLING

+ development of a single universal model far to ambitious (unrealistic)
+ multi scale approach is appropriate
+ closures for gas-particle and particle-particle interaction required
+ model should account for the transient nature of the flow

Binary mixtures: same size, different density: CARPT data available

New Delhi, India, September 2009

Tracking Solids in Gas-Solid Systems via Experiments and Modeling: RPT and DEM Studies

Rajesh K. Upadhyay, Ashish Abhinit , S. Vaishali & Shantanu Roy

Department of Chemical Engineering

Indian Institute of Technology (IIT) – Delhi

New Delhi, INDIA

Photo of 3-source X-ray scanner

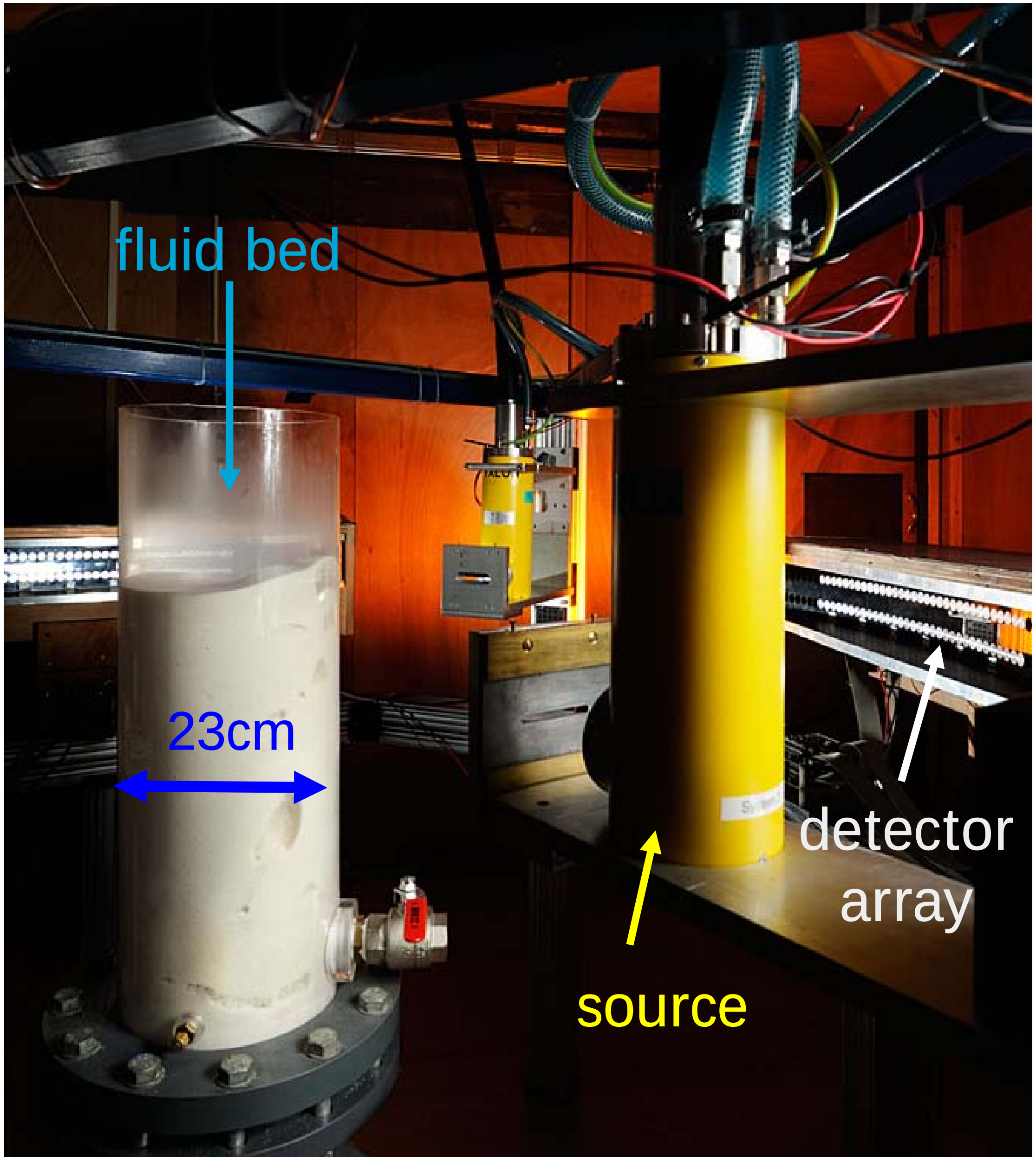


photo:
courtesy
Bart van
Overbeeke

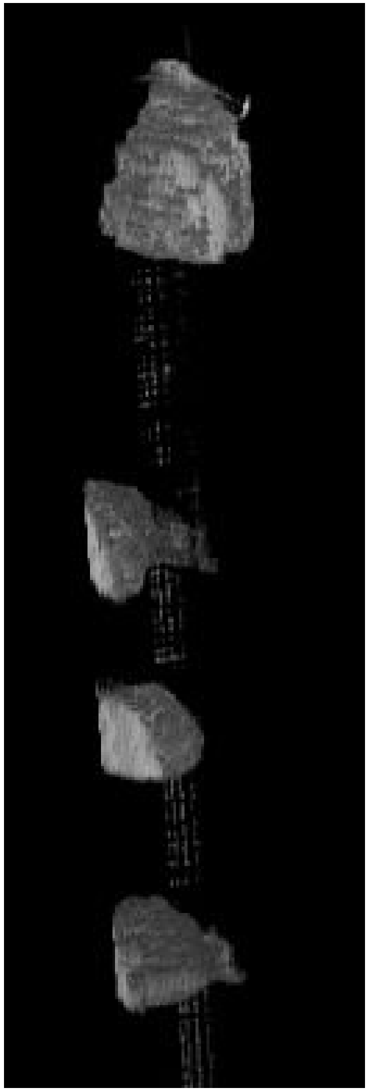
3D image of bubbling bed

- stack reconstructed images
- $\Delta t_{frame-frame}$: converted to distance

Usup~1.3*Umf

via **bubble velocity**

from time of flight
of each individual bubble
from lower to upper
detector plane

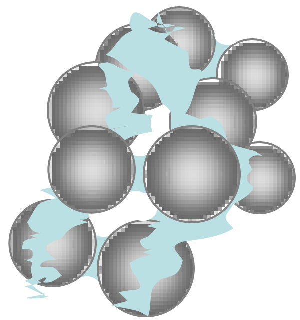


Usup~1.6*Umf

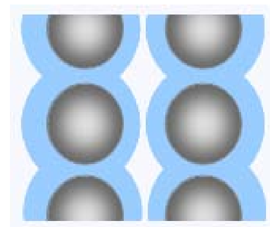
R. Mudde TUDelft

TBR Performance Assessment: Multi-Scale Approach

- TBR performance affected by particle scale & reactor scale flow phenomena
- Need to couple: 1) reactor scale CFD model; 2) particle scale models



"Rivulet flow"

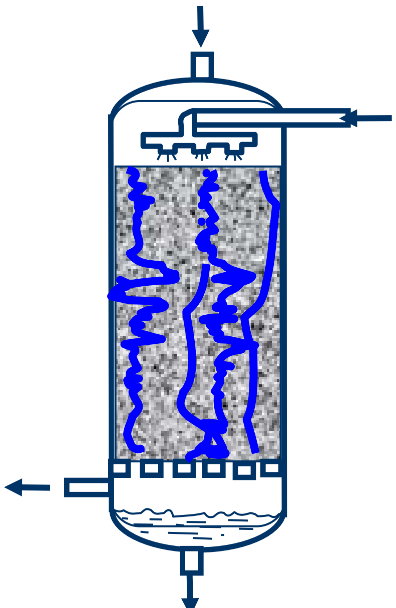


"Film flow"

Phenomenological analysis:

- Flow structures
- Description of phase interactions

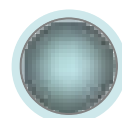
Can observe via NMR; Gladden et al.;,X-ray, Nicol et al.



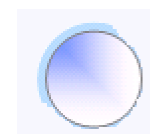
Reactor scale:

- Phase distributions
- Mal-distribution

Can observe via CT



Complete catalyst wetting

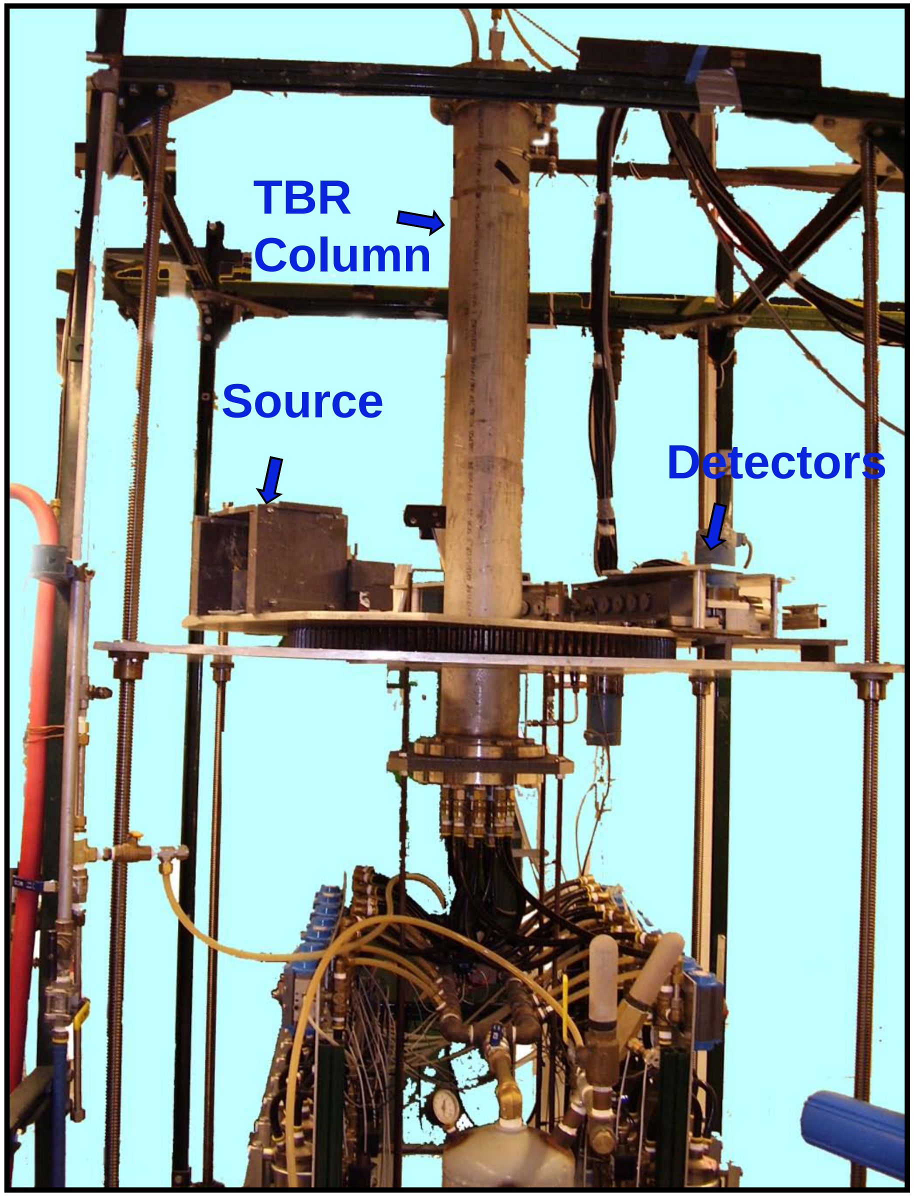


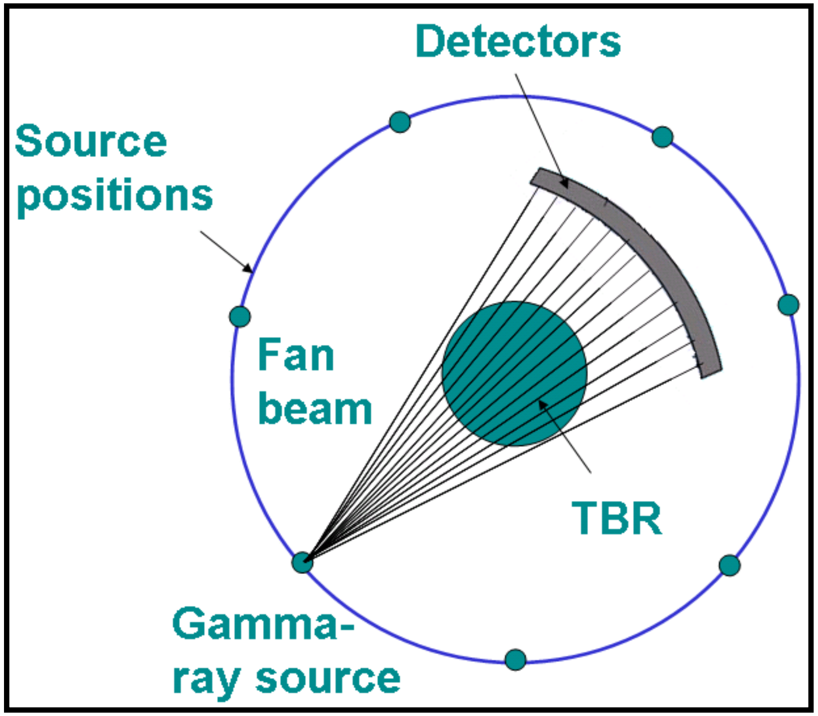
Incomplete wetting

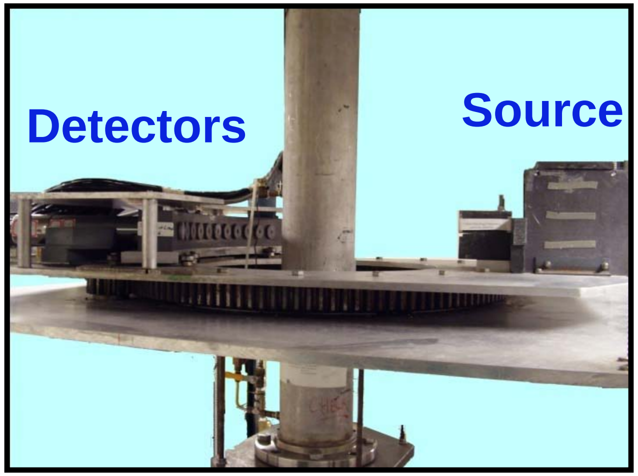
Particle scale model:

- Influence of local hydrodynamics
- Phase contacting and interphase mass/heat transport
- Intraparticle mass/heat transport (single or multi component) and reaction

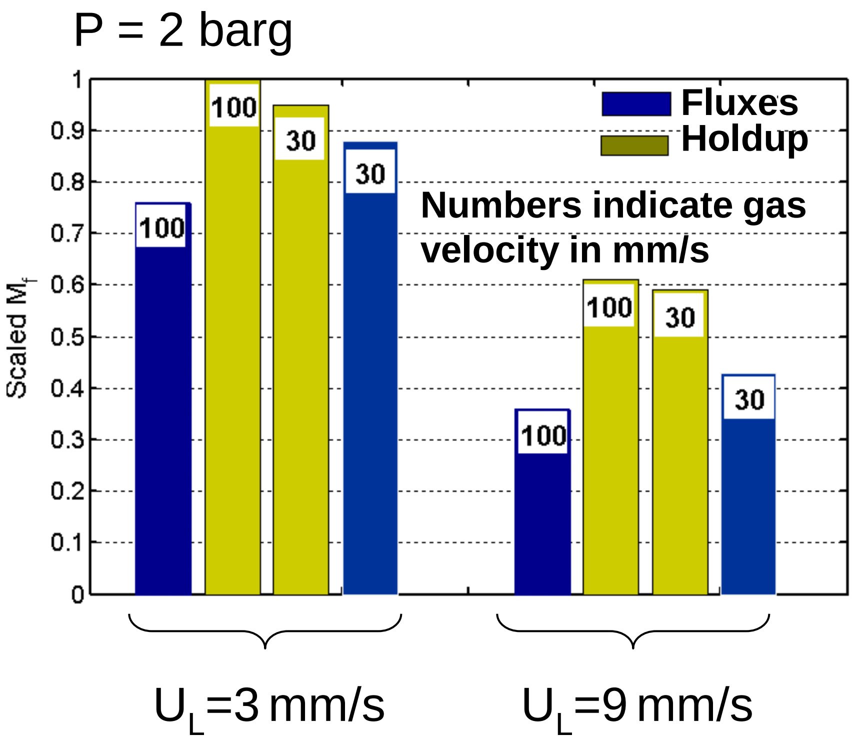
TBR and Computed Tomography (CT) Unit

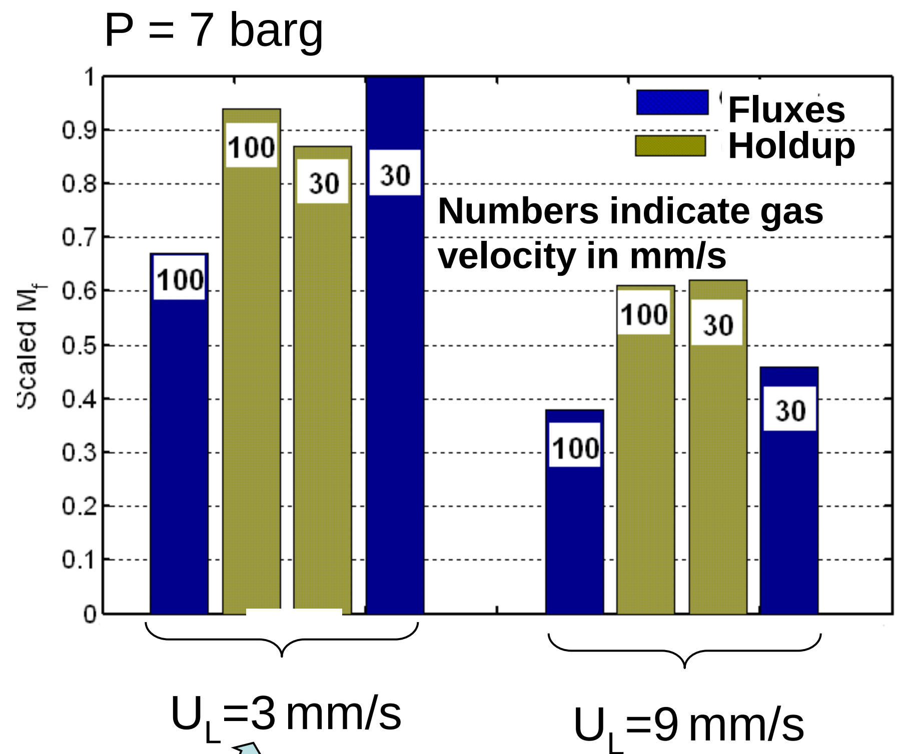






Fluxes and Holdup: Comparative Analysis





Mismatch between holdup and effluent fluxes maldistribution factor dependence on gas velocity

Maldistribution factor:

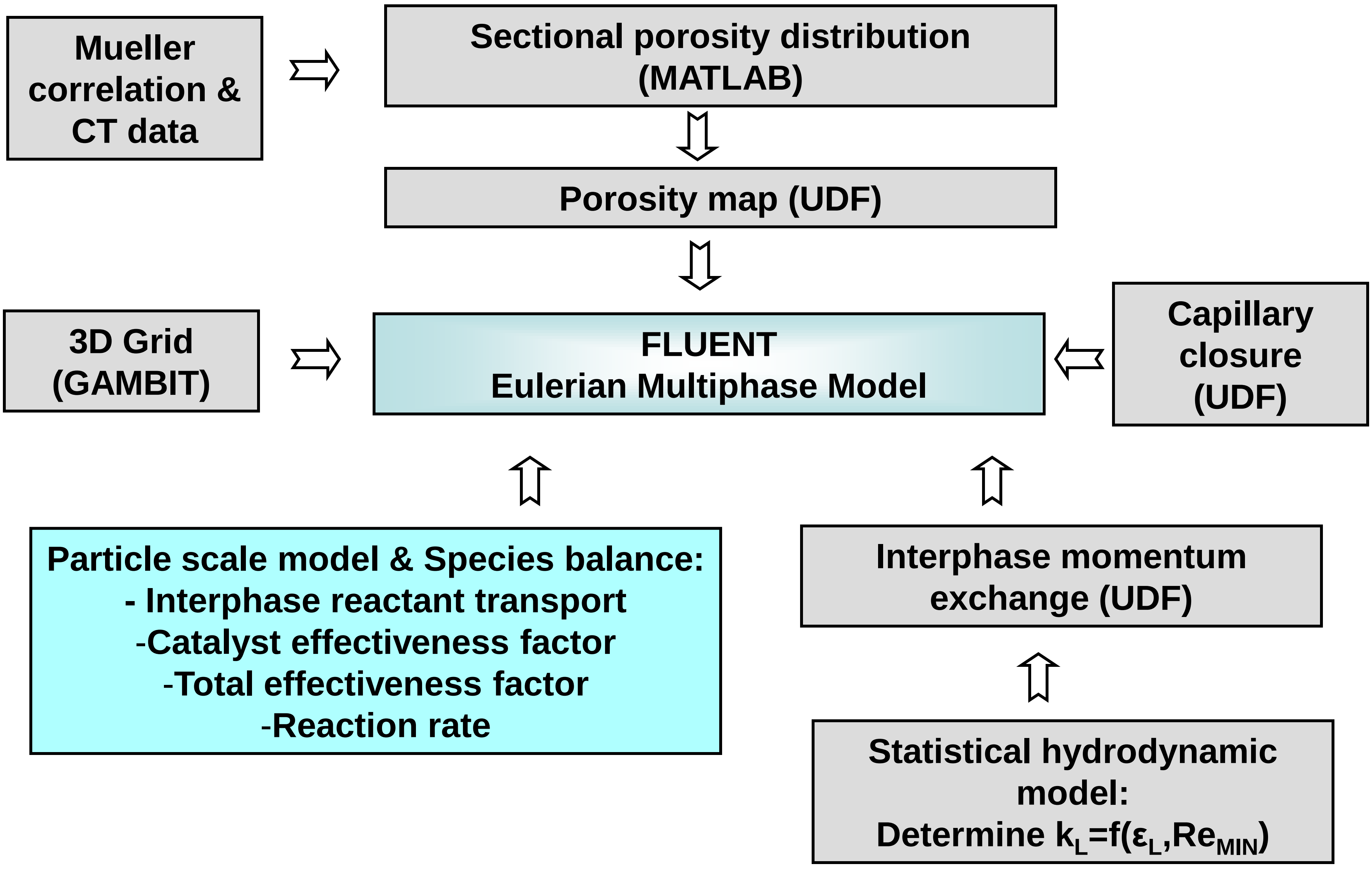
$$M_f = \sqrt{\frac{1}{N(N-1)}\sum_{i=1}^{N}\left(\frac{FLUX_i - \overline{FLUX}}{\overline{FLUX}}\right)^2}$$

Scaled M_f:

$$\left(M_{f,i}\right)_{scaled} = \frac{M_{f,i}}{\max\{M_{f,i}\}}$$

N = 15 (number of compartments)

Eulerian CFD Model Overview and Solution Procedure



3D Hydrodynamic Eulerian CFD Model

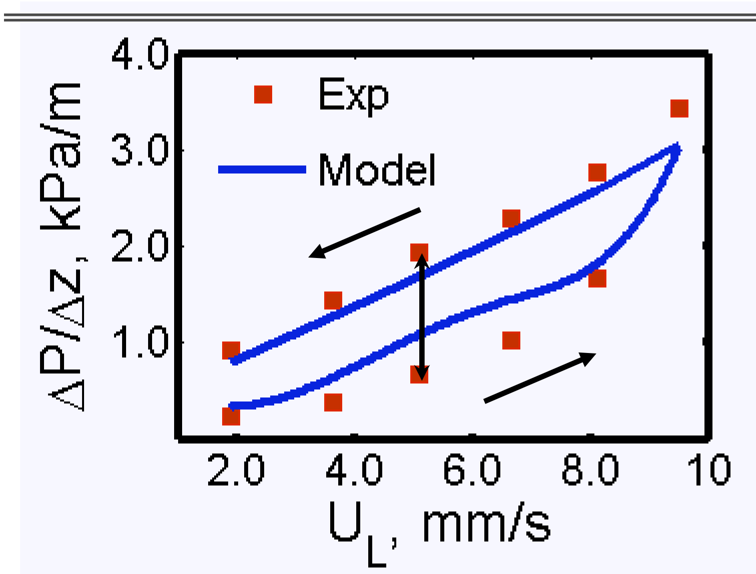
Model setup:

- Volume averaged equations on the computational grid
- Porosity distribution on the computational grid (CT data; Gaussian)
- Phase interactions closures (two fluid model, statistical hydrodynamics and relative permeability model)
- Account for pressure difference between gas and liquid phase (“capillary closure”)
- Solution strategy (Fluent/Gambit with Matlab and C codes)

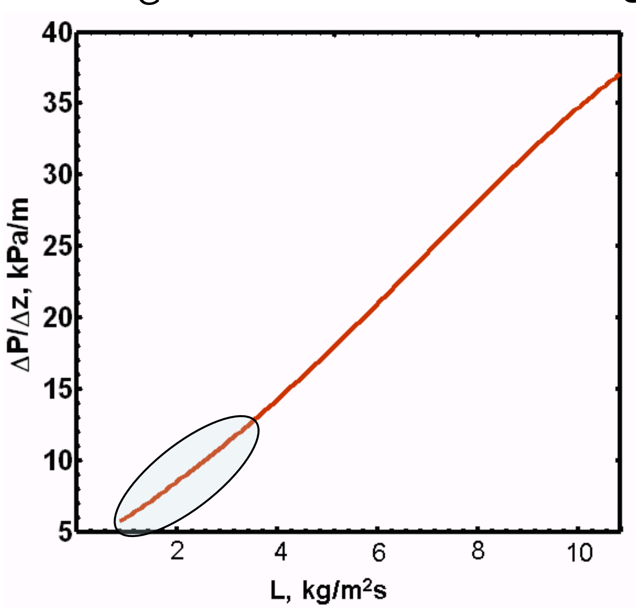
Basic input parameters:

- Ergun parameters (E_1, E_2 for the bed on interest via one phase flow experiments)
- Contact angle (determines likelihood of film vs. rivulet flow)
- Liquid phase relative permeability

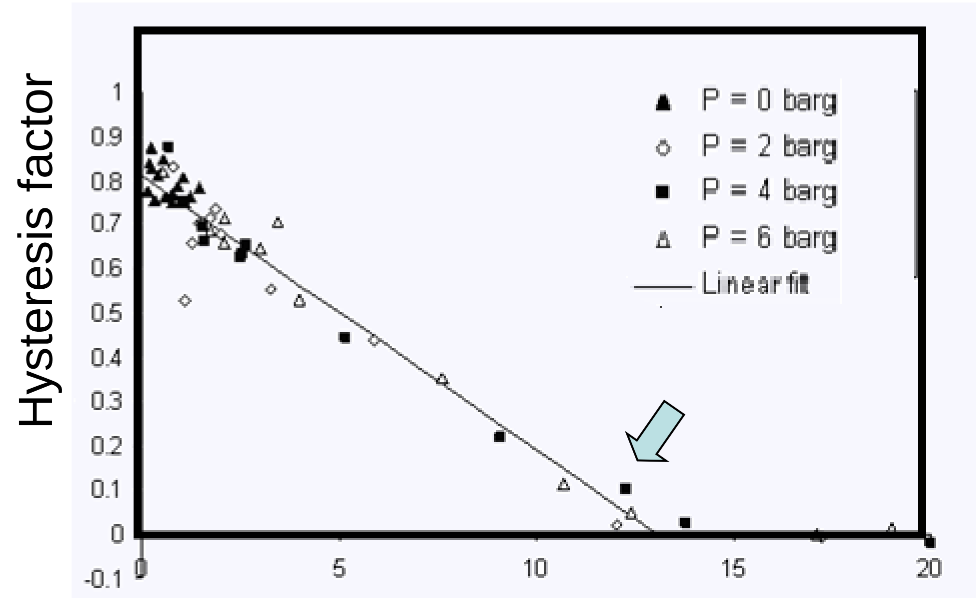
Predictions: Extent of Hysteresis



U_G = 27 mm/s, P=1 barg



Predicted pressure drop in Levec mode for conditions of HDS



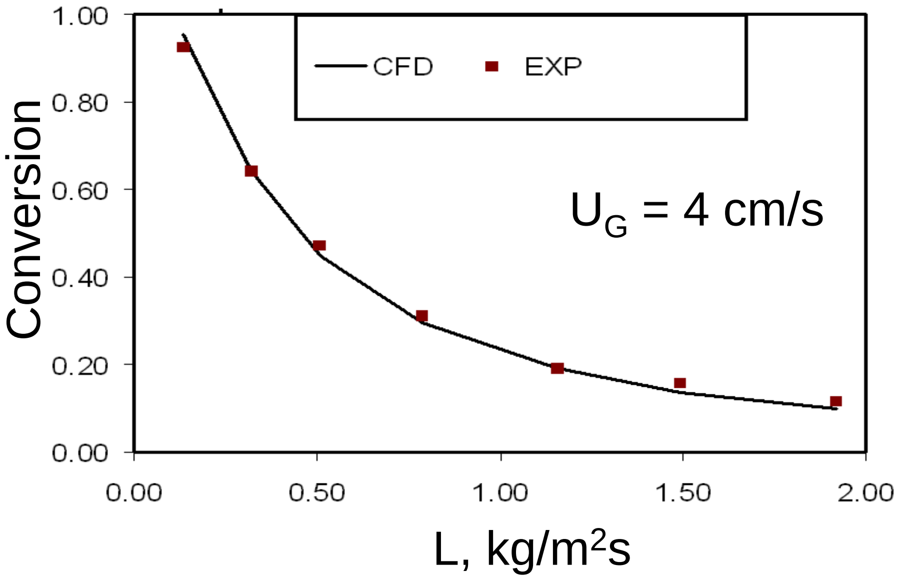
Experimental pressure drop in Levec mode, kPa/m

Levec prewetting mode:

Flood the bed; drain, and then initiate gas and liquid flow

$$f_H = 1 - \frac{(\Delta P / L)_{\text{Lower branch}}}{(\Delta P / L)_{\text{Upper branch}}}$$

Comparison with Exp Data*: Gas Limited



Hydrogenation of α-methylstyrene (to cumene) in hexane

$$\frac{C_L D_{eff,L}}{C_G D_{eff,G}} >> 1$$

- 1/16" 0.5% Pd on alumina

- Wetting Efficiency (El-Hisnawi, 1981):

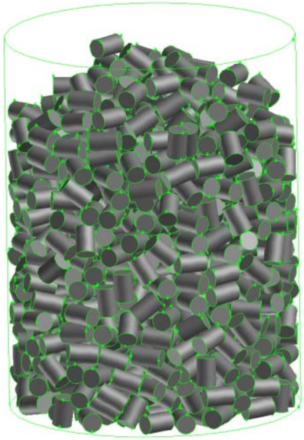
$\eta_{CE} = 1.617 \, \mathrm{Re}_L^{0.146} \, Ga_L^{-0.071}$ (Locally predicted by CFD model)

$$\eta = (1 - \eta_{\mathrm{CE}})\eta_{\mathrm{dry}} + 2\,(1 - \eta_{\mathrm{CE}})\eta_{\mathrm{CE}}\eta_{\text{half-wetted}} + \eta_{\mathrm{CE}}^2 \eta_{\text{fully-wetted}}$$

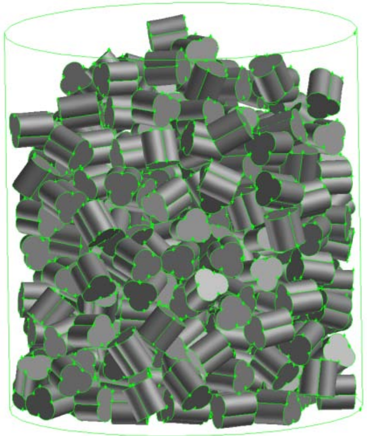
(Beaudry, 1987 model)

* *Experimental data of Mills et al., 1984*

Further Improvement in TBR Model by Micro-Scale Modeling of Packed Beds



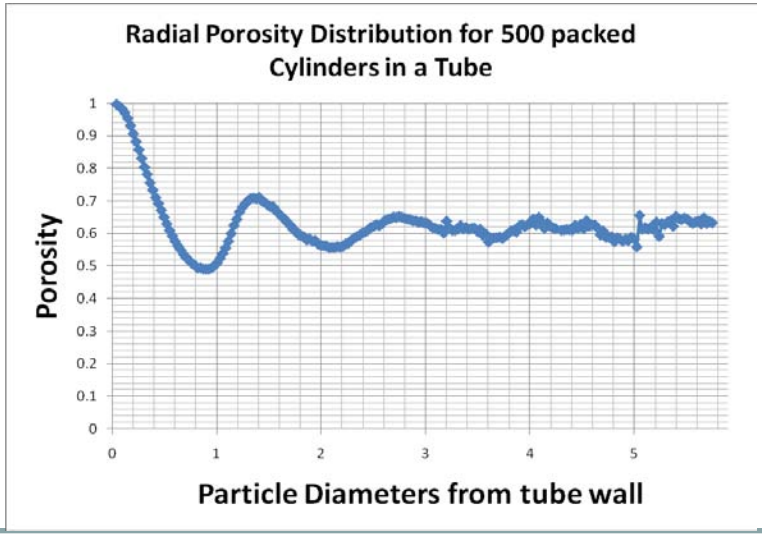
1000 Cylinders



250 Trilobes

- New Monte-Carlo packing algorithm makes producing random domains of cylindrical based particles possible.

- Simulations include complete local scale of catalyst particles modeled with Navier-Stokes equations explicitly.

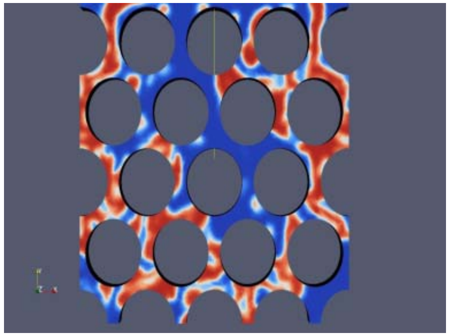


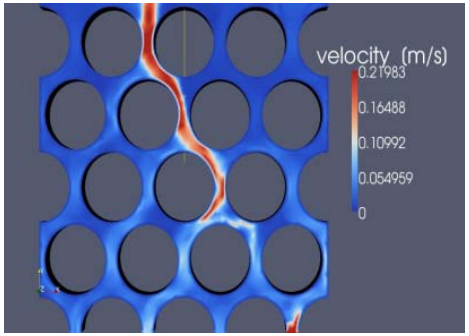
- Packed beds are loosely packed and can produce courser meshes than tightly packed beds

- Radial porosity distributions are comparable to those seen experimentally

Micro-Scale Modeling of Packed Beds

2D Trickle-Bed Flow Results (t=9.75 sec)
Gamma* [volume fraction] (left) and Velocity (right) Distributions**

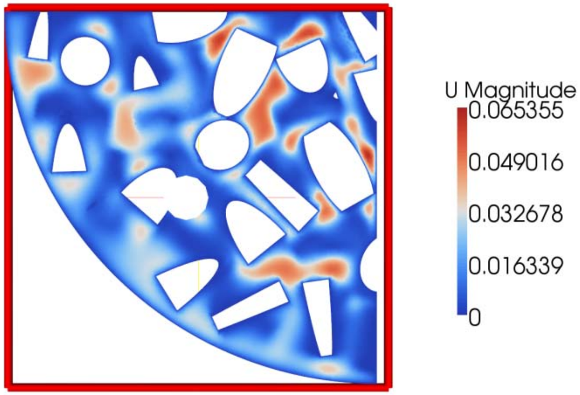




***In gamma results, red = liquid and blue = gas**

****higher velocity in red, lower in blue**

- 2-D multiphase flows are modeled with the Volume of Fluid (VOF) method.
- 3-D VOF results are difficult to obtain due to computational requirements



Horizontal cross-section
Z-Velocity Profile

- 3D single phase flows with turbulence are beginning to be available
- Key feature is that no Ergun-type pressure closure relations necessary for modeling.
- More advanced models incorporating heat and mass transport within the catalyst particle are possible using a coupled matrix approach that includes intra-particle transport.

Micro-Scale Modeling of Packed Beds

Packing

- Randomly packed domains of (10^2-10^3) cylindrical particles are made from a Monte-Carlo packing algorithm.
- Because the exact location of the faces of the particles are known, computational meshes are accurately constructed.

Modeling

- Micro-scale models of packed beds are based on the Navier-Stokes equations without an Ergun-type pressure closure relation
- More complex conjugate heat transfer models (including heat transport in solids) are being developed

Advanced Hardware Integration

- Because of the size of the sparse matrices produced by these meshes, a computational paradigm shift is necessary to leverage new technology in widely used CFD software.
- Integration of Graphics Processing Units (GPU) to solve these sparse linear systems is being performed with multiple times speedup compared to CPU based linear system solvers.
- Integration of GPU based solvers into OpenFOAM code is currently available.

Way Forward For Multiphase Reactors

- **Develop better multi-scale models and modeling framework to bridge the gap**
  - Essential to use multiple models to understand processes on different scales and to develop a framework to establish communication & data exchange among these multiple models
  - Experiments providing quality data are needed to discriminate & improve available models on all scales

- **Need to take two-track approach of pushing application envelope + developing new models**

  Get across clearly the message that computational modeling when validated provides invaluable support to engineering decision making & helps performance enhancement

To speed up the development we need a paradigm shift

- **Open source software**
  - Call upon an enormous software community for development of alternatives to traditional closed source commercial software
    - Linux, Firefox, VTK and so on
  - Source code is available to users
  - Open design for customization
  - Recent consolidations in commercial market leading to increasing acceptance of open source CFD
  - OpenFOAM: a leading open source CFD platform

Actions Needed to Facilitate Increased Application of MFS in Multiphase Reaction Engineering for Clean Technologies

Introduce multiphase flow and reaction engineering concepts in undergraduate and graduate curricula

Recreate or create with federal funding

-*IMUST* -*I*nstitute for *MU*lti-phase *S*cience and *T*echnology

with regular workshops for exchange of ideas and results and validation of codes

- MSPEF Multi-Scale Physics Experimental Facilities

at National Labs and/or Universities dedicated to study and visualization of multi-scale phenomena in multiphase flows and validation of the codes

Establish long term research targets for technologies of large environmental impact

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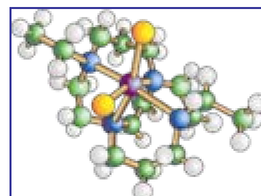

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